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Bench Scale Evaluation of Soil Washing for Heavy Metal Contaminated Soil at a Former Manufactured Gas Plant Site

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Soil contamination by heavy metals is a problem of great concern in all industrialized countries, where approximately 70% of the contaminated sites are polluted also by heavy metals. The most commonly found are lead, arsenic, chromium and zinc (US EPA 1996). The extent of contamination depends not only on the rate of loading, but also on the nature of interactions with adsorbing surfaces in the soil environment (Selim and Sparks 2001). Different kinds of technologies are available for the remediation of heavy metal contaminated sites. Among the most used technologies in Europe, soil washing appears particularly promising and with acceptable level of costs (Van Benschoten et al. 1997; Grasso et al. 1997). The term soil washing is used to describe a technology that either separates soil into particle size fractions using water, or chemically extract the contaminants from the soil. When no chemical is used to promote contaminant extraction from soil, the technology is strictly a physical process, in which finer soil particles are scrubbed from coarser, thus washing these last (Griffiths 1995). Physical soil washing is volume reduction treatment based on the intimate vigorous mixing of excavated soil with a solution, typically water, followed by a classification step, which separates soil slurry into different size fractions and isolate polluted particles smaller than a cut off size from uncontaminated coarser particles. It is based on the tendency of contaminants to adhere to soil fine particles, which, in turn, are attached to coarse fractions through forces of adhesion and compaction and can be removed by abrasive scouring and scrubbing action (Anderson 1993). This technology is aimed to concentrate the heavy metals into a reduced soil mass (typically 5 to 30% of the original soil volume) for subsequent treatment, whereas clean or slightly polluted soil can be returned to the site as fill, or otherwise used. Physical soil washing, which can be used also to treat radionuclide contaminated sites (Richardson et.al. 1999), is practicable when metals are mostly present as water insoluble forms. In this work a bench-scale treatability test was performed, for evaluating the applicability of this technology to soils at a former Manufacturing Gas Plants (MGP) site polluted with arsenic, zinc and lead.

MATERIALS AND METHODS

The investigated site: Officina Gas Bovisa, occupying approximately 430000 m^2 has been utilized for over eighty years (1908–1994) for the production of gas for the city of Milano (Lombardia, Italy) by coal gasification and catalytic reforming

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processes of petroleum light derivatives. The practice of manufacturing gas produced large quantities of wastes, among which arsenic and heavy metals (lead and zinc), as residues of the different processes of gas production.
Soil contamination by heavy metals was discovered in many areas of the site. Four "hot spot" zones (Table 1) were selected to ensure that the treatability test would be performed on samples representative of the soils with the highest contamination. Soil samples were collected at various depths: I10 ,3-4 m; I16, 0-1 m; I4, 4-5 m; 11SS, 0-1 m.

Mean soil characteristics of the different samples were very similar with a large percentage of sand (49%) and gravel (19%) a low content of clay (4%) and organic matter (1.9%), C.E.C. 12.2 meq/100 g soil, and pH values ranging from 7.2 to 7.4.

To use soil washing as a physical process with water as the liquid phase it is essential to characterize the heavy metal solubility. Solubility test was carried out according to the procedure UNI ENV 12506 reported in the Italian legislation for waste characterization (Italian Ministerial Decree D.M.5/2/1998). Soil samples are maintained under agitation in distilled water for 16 days with a ratio soil: water of 1:5 at 20±1 °C. Eight elution steps were performed by changing water after 2, 8, 24, 48, 72, 102, 168 and 384 h. In all the collected solutions heavy metals were determined by atomic absorption spectroscopy (Varian Spectra AA250 plus Furnace GTA97).

The bench-scale soil washing test was performed on 1 Kg of each soil samples selected. Samples were subdivided into sub-samples of 250 g (oversize coarser particles > 1 cm were discarded) and submitted to these following steps:
1) Water was added to contaminated soil sample in a ratio liquid to solid (L/S) 3:1 in 2L plastic containers to make a slurry. Preliminary experiments showed that the increase of L/S from 3 to 10 did not change the results significantly. Very low amounts of floating materials were observed and skimmed off.
2) The slurry was shaken at 20°C overnight in a high-speed agitator to facilitate the intense scrubbing of the soil particles. In this step the fines are removed from coarser size particles.
3) The slurry was fed after that to a screening process throughout different sieves in order to separate soil particles dimensionally. The following fractions were obtained: > 5 mm (i.e. between 10 and 5 mm), 5 - 2 mm, 2 - 0.2 mm, 0.2 – 0.1 mm, 0.1 – 0.05 mm, < 0.05 mm. The remaining soil material on each sieve was repeatedly washed with water until passage of finer particles through the sieve was not observed. The washing waters were recovered and passed on the following sieves with smaller mesh.

Each fraction was collected, oven dried at 105°C, and weighted. After acid digestion, arsenic, lead and zinc were determined by atomic absorption spectroscopy (SSSA 1996). All waters from the process were also analysed for the same elements.

Table 1. Heavy metal concentration in the four unwashed soils. Data (mean ± SD) are expressed as mg Kg-1 on a dry weight basis (105°C).

Metals	Soil			
	I 10	I 16	I 4	11 SS
Lead	3625 ± 111	600 ± 28	750 ± 41	245 ± 22
Zinc	2975 ± 204	250 ± 36	275 ± 38	146 ± 18
Arsenic	10.3 ± 2.4	276 ± 45	67 ± 9.0	700 ± 54

RESULTS AND DISCUSSION

Heavy metals are retained by soil through processes of sorption, ion exchange, precipitation – solubilization, complexation, etc.. These reactions produce a wide spectrum of interactions among heavy metals and soil components: humic substances and minerals, characterized by linkages of different energies.

The key to evaluate if a contaminated soil is amenable to physical soil washing is the characterization of these soil contaminants relationships, in particular solubility and particle size distribution.

Data from the solubility test showed that heavy metal concentrations were negligible in all the eight water extracts obtained. These results strongly address the use of water in the washing process.
The alternative: solubilization of metals in the liquid phase (chemical soil washing) was discarded because:
a) a remarkable variation in the composition of the liquid phase of the washing process would be necessary to solubilize the metals with meaningful extraction efficiencies (for example strong acidification).
b) the chemical behaviours of Pb and As show opposite solubility trends at varying pH. A two-step process would be required to solubilize both elements and it would be necessary a further treatment of washing waters with a consequent increase of the costs.

Physical soil washing requires that a high proportion of soil particles would be of coarser dimensions. Following the washing treatability test used, soil was subdivided into fractions of different sizes. Weight distribution of the different particle size fractions was nearly homogeneous in the four investigated soils; results are reported in table 2. The size fraction > 5 mm was the predominant in all the samples with a value around the 55% of the total soil weight, while the size fractions comprised between 0.2 and 5 mm accounts from 25 to 30% of the total soil. The fraction between 0.1 and 0.05 mm was the less relevant from 2.5% to 3% of the total soil weight. Finally the fraction below 0.05 mm accounted for about 10% of the total soil.

Table 2. Percentage of weight distribution of the different particle size fractions of soils.

Size fraction (mm)	Soil			
	I 16	I 10	I 4	11 SS
10 - 5	52.8	55.6	53.7	54.0
5 - 2	9.38	11.1	11.8	12.0
2 – 0.2	20.1	16.7	17.6	17.4
0.2 – 0.1	2.1	2.8	3.4	3.1
0.1 – 0.05	2.6	2.6	2.1	3.0
< 0.05	13.0	11.2	11.4	10.5

The goal of the treatability test was to determine if target levels of concentration for the site could be achieved from particle size separation using water alone.
The distribution of Pb, As and Zn concentration in the different size fractions of the soils, following the bench scale test, is reported in figures 1,2,3 and compared with the remediation goals (dotted lines) deriving from the site-specific risk analysis (Bonomo et al. 2000). There is no remediation limit for Zn, being an essential microelement, not particularly toxic.

Results showed that the trend of distribution of contaminants is the same for all soils: the concentration increased as decreasing the particle sizes. In all samples the washing process reduced the concentration of heavy metals in the coarser fractions, even if differences were found among soil samples and contaminants. Concerning lead, the concentration in the coarser fractions (> 5mm and >2 mm) is below the remediation limits, in all soil samples (fig.1). The washing process is particularly efficient for samples I 4 and I 16 starting from concentration not too higher than the remediation limit. The washing process was also high effective in removing lead form coarser particles in the soil sample I 10 characterized by a very high initial concentration. In all soil samples the fraction > 5 mm contain less than 1 mg/kg of lead, while the fraction between 2 and 5 mm showed a concentration ranging from 25.0 mg/kg (I 16) to 165 mg/kg (I 10).

Arsenic concentration in the different soil fractions showed a similar trend (fig. 2), and the clean-up goal is reached in all soil samples for the fraction >5mm, with a concentration below 1 mg/kg. Positive results were obtained also for the fraction 2–5 mm in soil samples I 10 and I 4 with a concentration below 8 mg/kg for both soils. Similar result seems difficult to obtain for samples I 16 and 11 SS, which contained after the washing process 67 mg/kg and 330 mg/kg respectively.

The trend of Zn concentrations in the different size fractions of soils is similar to the one of Pb and As (fig. 3). For all soil samples the concentration of the metal is less than 1 mg/kg in the fraction >5mm and range from 4 mg/kg to about 300 mg/kg in the fraction 2-5 mm. These values of concentration are quite acceptable being 300 mg/kg the soil limit value for the utilization of sewage sludge in agriculture (EC Directive 1986).

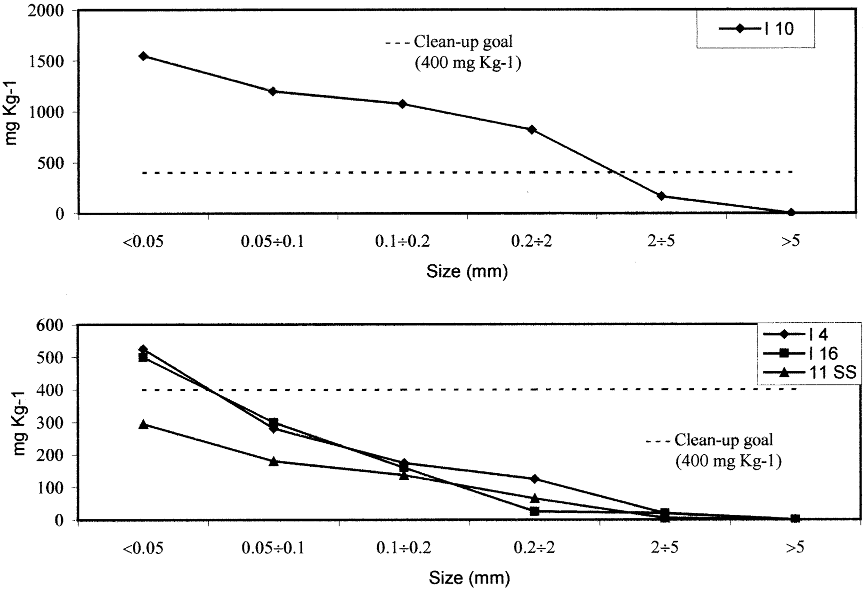

Figure 1. Distribution of Pb concentration in the different size fraction from the soil washing process.

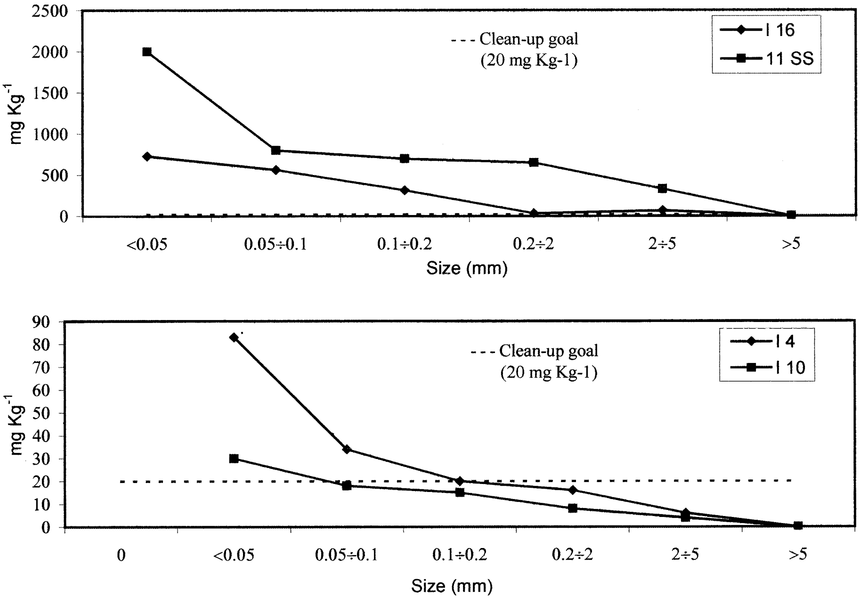

Figure 2. Distribution of As concentration in the different size fraction from the soil washing process

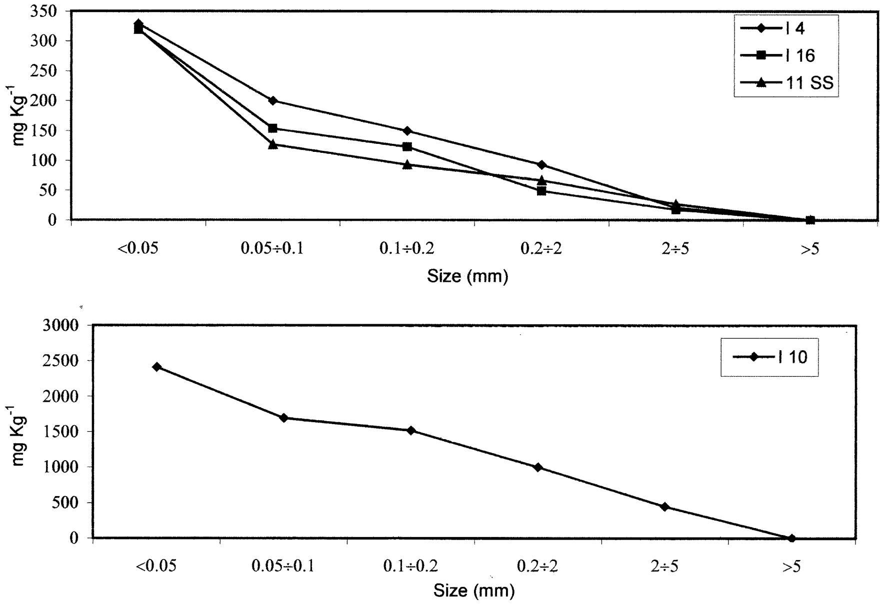

Figure 3. Distribution of Zn concentration in the different size fraction from the soil washing process.

It is, however, reasonable to infer that the levels of concentration in the coarser size fractions will be further reduced, by blending soils from different zones with lower concentration before the washing treatment, since the collected soil samples derived from "hot spot" zones. A further reduction in concentration could be presumably obtainable in a full-scale process in which the scrubbing conditions are drastically more vigorous.

The efficiency of the process was evaluated by means of contaminants removal and potential volume reduction. Soil washing is considered a viable technology if the reduction of contaminants concentration in the fraction > 2 mm allows a recovery of, at least, 50% of total soil as clean soil (Anderson 1993, Pensaert 2003). Obtained data (Table 3), pointed out that physical soil washing can be successfully applied in this site; the better results were obtained for lead with an efficiency of more than 90%, whereas a lower efficiency was found for As ranging from 52 to 89 %.

The finer fractions, characterized by the highest contamination, account for about 15 - 20% of the total soil, so that about 80% of the original soil can be reused, without further treatments. A further improvement of the efficiency of the process might be achieved in the full-scale process where remarkable forces of friction are involved much more vigorous than in the laboratory test.

Table 3. Efficiency of soil washing considering the >2 mm fraction only. Data are expressed as percent of metal removal in the respect to the total concentration in the original unwashed soil.

Metal	Soil			
	I 10	I 16	I4	11 SS
Pb	96.6%	95.4 %	92.7 %	97.9 %
Zn	84.1 %	94.0 %	92.7 %	84.5 %
As	85.7 %	63.0 %	89.1 %	52.8 %

In conclusion this bench-scale study provides a detailed knowledge of distribution of heavy metals in the various dimensional classes of the soil particles, which is of great help for a sound forecast of the results obtainable by the technology. Moreover obtained data will be useful for the potential soil washing contractors to assess the technical requirements and the related costs.

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REFERENCES

Anderson WC (1993) Soil washing/ soil flushing. Springer Verlag Heidelberg

Bonomo L, Caserini S, Pozzi C, Uguccioni DA (2000) Target cleanup levels at the site of a former manufactured gas plant in Northern Italy: deterministic versus probabilistic results. Environ Sci Technol 34:3843-3848

European Community (1986) EC Sewage Sludge Directive 86/278/EEC

Grasso D, Butkus MA, O'Sullivan D, Nikolaidis NP (1997) Soil washing design methodology for a lead contaminated sandy soil. Wat Res 31:3045–3056

Griffith RA (1995) Soil washing technology and practice. J Haz Mat 40:175-189

Italian Ministerial Decree D. M. (1998) Individuazione dei rifiuti non pericolosi sottoposti alle procedure semplificate di recupero ai sensi degli art. 31 e 33 del D. Lgs. 5 /2/ 1997 n° 22

Mann MJ (1999) Full scale and pilot scale soil washing. J Haz Mat 66:119–136

Pensaert S (2003) The remediation of Sir John Rogerson's quay gasworks at Dublin: a case study. In 8th International FZK/TNO Conference on Contaminated Soil (Consoil 2003), Gent, Belgium. Theme C 1896-1905

Richardson SW, Phillips CR, Luttrell J, Hicks R, Cox C (1999) Application of remedy studies to the development of a soil washing pilot plant that uses mineral processing technology: a practical experience. J Haz Mat 66:47–65

Selim HM and Sparks DL (2001) Heavy metals release in soil Lewis Publisher CRC Press Boca Raton USA

SSSA Book Series: 5. Methods of soil analysis Part 3. Chemical Methods. Soil Science Society of America Inc. Madison Wisconsin (1996)

USEPA (1996) Report: Recent developments for in situ treatment of metals-contaminated soils. Office of Solid Wastes and Emergency Response, U.S.Environmental Protection Agency, Washington DC

Van Benschoten JE, Matsumoto MR, Young W (1997) Evaluation and analysis of soil washing for seven lead contaminated soils. J Environ Eng 123:217–224