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Development, assessment and evaluation of a biopile for hydrocarbons soil remediation

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ABSTRACT

Soil at a site located in North-Eastern Italy has been impacted by a persistent, long term exposure to diesel fuel. The concentration of organic contaminants in soil exceeded the acceptable limits indicated by the present regulations concerning the specific use-destination of the sites and its reclamation. The contamination involved several thousands square meters of soil surface resulting in some thousand cubic meters of soil. Approximately 650 cubic meter of the most contaminated soil was moved to a nearby site where a biopile was assessed with the aim to enhance the activities of the soil community to reduce the oil concentration. In order to verify the correct operation of the system it was continuously monitored in terms of chemical concentration, biological activity and community structure. The main remediation occurred within the first 300 days of treatment allowing the soil to be back within the threshold defined by current laws for industrial and commercial activities. An additional 200 days treatment was needed to further reduce the oil content to the limits required for public parks and residential areas. Lab scale biopiles were also assessed to evaluate the role of commercial inoculants in the remediation process.

Introduction

Hydrocarbons represent one of the most common contaminants requiring remediation due to the problems they pose to human health and controlled waters (Ollis, 1992; Kirk et al., 2004). Hydrocarbon compounds derive from petroleum sources, including common fuels such as gasoline, diesel, kerosene and lubricating oils and greases. Although hydrocarbons are organic substances composed of only carbon and hydrogen, they include a vast number of different compounds each revealing distinct chemical and physical properties. Therefore, the remediation methods to be applied need to be evaluated each time, on a site-specific basis and in accordance with regulatory agency guidelines (Alexander, 1995; Jørgensen et al., 2000; Khan et al., 2004).

Biopiles, also known as biocells, bioheaps, biomounds, and compost piles, represent a reliable system to reduce the concentration of petroleum constituents in excavated soils through biodegradation (Genovese et al., 2008). This technology involves collecting and transferring contaminated soils to a dedicated site specifically set up to stimulate aerobic microbial activity within the soils through the aeration and/or addition of minerals, nutrients, and moisture. The enhanced microbial respiration results in degradation of adsorbed petroleum-product constituents (Thompson et al., 2005).

In this work the "biopile approach" was applied to a soil of North-Eastern Italy (46°01′15″ N, 11°54′01″ E; elevation above sea level: 275 m) contaminated by a diesel fuel spill for a period of 20–25 years, as revealed by workers starting the construction of a new building. The hydrocarbon concentration in soil significantly exceeded the acceptable limits indicated by the present regulations concerning the specific use-destination of the sites and its reclamation. The contamination affected a surface of several thousand square meters resulting in some thousand m^3 of polluted soil and the high concentration of oil found in this area caused local authorities to stop the ongoing construction project and determine the most effective means of remediating soil from the site.

Analysis of the underground water from one of the piezometers installed during a preliminary phase, showed a gas chromatographic profile characteristic of a high density fuel oil (ARPAV,

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Agenzia Regionale per la Prevenzione e Protezione Ambientale del Veneto, personal communication). The dominant mechanism to break down the non-volatile petroleum components of heavier petroleum products (e.g., heating oil, lubricating oils) is biodegradation. Moreover, higher molecular weight petroleum constituents such as those found in heating and lubricating oils, and, to a lesser extent, in diesel fuel and kerosene, require a longer period of time to degrade than do the constituents in gasoline (Khan et al., 2004). All this strongly suggested the preparation of a suitable biopile at a nearby site would be the most effective treatment method. In order to verify the correct operation of the system the plant was continuously monitored in terms of concentration of contaminants and presence and activity of several microbiological groups and all the data collected are here reported. Since a commercial inoculant was applied in the bioremediation process, additional lab-scale biopiles were assessed with the aim to evaluate the possible role of the newly added microbiota.

Materials and methods

Arrangement of the biopile

Approximately 650 cubic meter of soil was selected as the most contaminated fraction and transferred to a nearby site where a biopile was constructed by Serenambiente Srl (Villa Agnedo, Trento, Italy) with the aim to speed the activities of soil microorganisms in reducing oil concentration.

To make the system completely confined, the engineered bioremediation structure, 23 × 14 × 2 m, was assembled by arranging a fine sand mattress with geotextile and HDPE (high-density polyethylene). An aeration system was provided and the soil moisture was maintained by re-circulating water amended with nutrients. Since it was unworkable to have available an entire control biopile, lab scale versions of the system were also assessed for suitable comparisons. In the main biopile, and in all these separate small versions, the wetting system could also be used to periodically release a commercial microbial inoculant containing a dried consortium of bacterial strains, declared as effective by the manufacturer for hydrocarbons degradation. The inoculant, contained $9 \cdot 10^9$ microorganism$\cdot g^{-1}$mixed with a bentonite-clay-based support, but the specific chemical and microbial composition of the product is proprietary.

Experimental design

The biopile was operated for more than 500 days during which time analyses were performed on soil samples taken from various sites. Since the soil of the biopile showed some evident heterogeneity in the oil distribution, a theoretical subdivision of the biopile into 8 different sub-plots (A1, A2, B1, B2, C1, C2, D1, D2) was designed. Chemical analysis were undertaken from the beginning of the experiment (T0: May 13) and after 7, 21, 33, 38, 47, 54, 150, 327, 337, 372, 407, 440, 472, 512 and 540 days, while microbiological and molecular analysis were performed on plots A1, B1, C2 and D2.

As natural controls for the main biopile, additional soil samples were collected and analysed from the polluted (P-Control) and unpolluted (NP-Control) soils surrounding the contaminated site since they did not receive the physical-chemical-microbiological soil treatments applied in terms of aeration and addition of minerals, nutrients, moisture, and inoculant. In addition, two separate small control biopiles were also assessed. These were designated as (i) BPI-Control, containing the same polluted soil collected at the experimental site, equipped with a wetting system and in which the commercial inoculant was constantly applied during the whole period, thus replicating the main biopile in a reduced version; (ii) BP-Control, equipped as the above BPI-Control, but where the commercial inoculant was not applied.

In order to obtain an extended evaluation of the effectiveness of the applied inoculant, two other small biopiles were assessed, containing a different, unpolluted soil, which was artificially contaminated with 3.65 ml/kg of diesel fuel (corresponding to the initial average level of contamination of the main biopile soil). These two lab-scale biopiles differed only in the application of the commercial inoculant and so were designated NSI-Control and NS-control, with the I sample having inoculant added.

Chemical analysis

For soil extraction, the protocol described in ISO/TR 11046/94 was followed. In short soil samples were ground, 2 mm sieved and dried in vials by addition of anhydrous sodium sulphate. After drying, extractions were performed with 1,1,2-trichloro-1,2,2-trifluoroethane (Freon 113) under sonication for 2 h (47 KHz, Ultrasonic Bath 1200, Branson Danbury, USA). The extract was transferred to a purification column (Florisil® 60–100 mesh, Sigma–Aldrich, St. Louis, USA) and the eluate completely collected. The purified extract was then stored at −20 °C in gastight vials until gas chromatographic analysis. Oil content was evaluated by analysing a range of hydrocarbons from C8 to C40.

At present, Italian legislation indicates that total hydrocarbons (C > 12 and C < 12) cannot be identified by a single analytical method that meets the criteria of specificity and selectivity and ISS (Istituto Superiore della Sanità) specifies the adoption of ISO/TR 11046/94. Total hydrocarbon analysis requires the specific compound that caused the contamination of soil as a standard. Since the pollution in the study here is from a more than 20 year old source the original fuel mix is unavailable. Therefore, it was considered appropriate to use certified standards (ULTRA Scientific, Kingstown, USA), for TRPH (Total Recoverable Petroleum Hydrocarbons: even n-alkanes C8 to C40) and DRO (Diesel Range Organic: all n-alkanes C10 to C28 with boiling temperature 170–430 °C) analyses. n-alkanes with C < 10 were evaluated by EPA method 8015b using certified standard (ULTRA Scientific, Kingstown, USA) for GRO (Gasoline Range Organic); VOCs (Volatile Organic compounds) were determined by GC–MS as reported by UNI 10899:2001.

Gas chromatographic analysis were performed adopting ISO/TR 11046/94 by a TRACE GC 2000 (Thermo Scientific, Waltman, USA) equipped with a VF-5ms column (Agilent Technologies, Santa Clara, USA) (30 m × 0.32 mm with 0.50 μm film) and a split injection ratio of 1:45 with helium as the carrier gas (1.8 ml/min). A FID unit was used with an operating temperature of 350 °C with an injection port temperature of 270 °C. The oven temperature was programmed 40 °C (3 min) to 340 °C (18 °C/min) and held at this temperature for 15 min. Total program length was 35 min.

The calibration was performed using the internal standard 5-alpha-androstane (ULTRA Scientific, Kingstown, USA); the interval of integration was between the retention time of n-decane (C10) to n-tetracontane (C40).

Microbiological analysis

Plate counts

The number of culturable microorganisms were determined by standard plate count method. Soil suspensions were prepared by vigorously shaking 20 g of soil in 180 ml 0.9% NaCl for 30 min. Then 100 μL of each dilution level was plated in quintuplicate onto Plate Count Agar (PCA) (HiMedia Lab., Bombay India) to enumerate aerobic heterotrophic micro-organisms. Diesel-resistant populations were quantified on PCA medium agar plates where the