



Note from the field

Remediation of contaminated soils by thermal desorption; effect of benzoyl peroxide addition



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ABSTRACT

In situ remediation of contaminated sites by thermal desorption is a widely accepted technology that provides a permanent solution at an economically competitive cost. In this paper, thermal desorption of various model contaminants was tested at the bench scale. Polyaromatic hydrocarbons (benzene/toluene/xylene) and non-polar extractable compounds were selected as representatives. The electrically heated static oven worked up to 380 °C with temperature gradient 6 °C/min, under air flow (0.6 m³/h). Prior to an experiment, model contaminants (xylene, 2-methylnaphthalene or motor oil) were mixed with 18 kg of model soil in concentrations 2–10 g/kg. The desorption process was performed with and without addition of 1 wt. % of benzoyl peroxide as an oxidant. The aim of the oxidant addition was to facilitate the chemical catalytic decomposition and/or biodegradation of desorbed organics from the soil as an alternative to usually employed incineration. In the course of the desorption process, part of the organic contaminants was oxygenated and liquids obtained after condensation of the desorbed contaminant vapours were subsequently treated by biodegradation. The application of benzoyl peroxide significantly accelerated biodegradation of organic compounds present in the condensates by *Pseudomonas* sp. Moreover, it was shown that the presence of partial oxidation products in the condensates did not increase their eco-toxicity.

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1. Introduction

Contamination of soils with oil and petroleum-based hydrocarbons in accidental spills is causing increased environmental concern. In many cases, remediation of contaminated sites by thermal desorption is the best option (Kastanek et al., 1994; Skvara et al., 2002; Khan et al., 2004). A wide range of remediation technologies based on thermal desorption exist (Kastanek and Kastanek, 2005; Mirsal, 2008; Pavel and Gavrilescu, 2008), as well as numerous EPA superfund studies (U.S. Environmental Protection Agency, 2000). A carrier gas or vacuum system transports volatilized water and organics to the gas treatment system, where the organics are destroyed in a secondary combustion chamber or in a catalytic reactor. A preferred alternative from an

environmental point of view is the direct destruction of vapours in an adjacent catalytic reactor.

However, little is known about the effect of water vapour on different oxidation reactions and a possible negative effect of water vapour on activity or selectivity of the catalyst such as Pt-Pd/Al₂O₃ and/or Cu-Mn/Al₂O₃ (Kikuchi et al., 2002; Matejova et al., 2012). To address the problem, partial condensation of soil moisture and organic vapour could be carried out in an intermediate stage between the desorption chamber and the catalytic unit. The condensed organic vapour, containing catalyst poisons like sulphur compounds or heavy metals, could then be handled separately, for example using biodegradation, whilst the remaining (uncondensed) vapour containing the organic components, which form aerosols and are difficult to condensate, can be destroyed in the catalytic reactor (Solcova et al., 2014).

Nevertheless, the biodegradation process is considerably slower owing to some of the xenobiotic organics, produced by modern industrial processes (e. g., polyaromatics), that resist biological treatment. Although the organic contaminants volatilize at typical

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soil temperatures and typical residence times used in current thermal desorption units, they are not oxidized. Therefore, following thermal desorption, it would be beneficial to obtain partially oxidized contaminants prior to their introduction into the subsequent biodegradation or catalytic oxidation stages (Matzavinos and Psillakis, 2004). This can be achieved using ozone, hydrogen peroxide or potassium permanganate, which are generally applied in liquid phase. Other well-known oxidants (Fenton, potassium permanganate) that have been previously applied in wet oxidation experiments introduce an excess of inorganic elements into the soil. Furthermore, hydrogen peroxide rapidly decomposes on contact with soil even at moderate temperatures and aeration of soil with ozone is technically difficult, especially when thermal desorption of the organic compounds is performed in stationary soil layers.

Consequently, in the work reported here, benzoyl peroxide was selected as a strong organic oxidant for which it was presumed that several oxidation steps take place in the course of thermal desorption in the presence of benzoyl peroxide and the target organics are partially oxygenated into simpler chemical substances prone to subsequent biodegradation and/or catalytic decomposition. However, given the potential for explosion of organic peroxides (Kozak et al., 2011), safeguards must be in place. Here dosing was kept up to 1 wt. % in soil, and under low heating rate (up to 6 °C/min), where the application of this oxidant was safe. Although various studies of benzoyl peroxide with organic compounds have been done in the past, they were mostly carried out as the free radical arylation reactions in nitrogen atmosphere. The application of oxidative additives in thermal desorption of organics from soil has not been studied yet.

Emphasis is now also placed on *in situ* methods like washing and/or drainage of contaminated soils. There is sometimes the problem concerning waterlogged contaminated soil (Valipour, 2014; Yannopoulos et al., 2015). Then, drainage of contaminated land is required to reduce waterlogging and soil salinization that inevitably accompanies waterlogging in arid zones similarly as to flood out the soil contamination, if any. One of the most important factors for appropriate performance of subsurface drainage systems is having adequate inflow to the drain for improvement of soil environmental conditions. For this purpose, knowledge about effect of drainage parameter's change on drain discharge is essential in subsurface drainage systems (Valipour, 2013a, 2013b).

The dewatering (drainage) by electrokinetic decontamination process is well known and it is commonly used for decades. However, numerous factors affect the soil decontamination by electrokinetic drainage and only some types of soils are suitable (Fang, 1997). Typical data for various types of soil decontamination by these method show that water quality before and after the drainage process has been efficiently improved mainly for alkalinity and organic (and ammonia) nitrogen removing. However, the data concerning the drainage efficiency of organic contaminants are scarce and more investigation is needed. The potential use of drainage processes of soil decontamination can be enhanced if the basis of mechanisms of soil-electricity interaction will be more elucidated. Nevertheless, drainage of contaminated soil can be considered as a conceivable preliminary step to the following excavation of contaminated soils and thermal desorption treatment.

The aim of this research was to verify the possible occurrence and composition of oxidized products in vapours when partial oxidation of selected hydrocarbons is carried out in the course of thermal desorption, following addition of an oxidizing agent into the treated soil. The point of the oxidant addition was to facilitate the chemical catalytic decomposition and/or biodegradation of

desorbed organics from the soil as an alternative to usually employed incineration. The addition of the oxidizing agent - benzoyl peroxide-into the treated soil is a new step in the process of thermal desorption, which ensures that the decontamination of pollutants in the soil and their destruction proceeds rapidly and completely. Furthermore, the rate of biodegradation of hydrocarbons and their partially oxidized products in shared aqueous condensates was evaluated.

2. Experimental

Thermal desorption of organic compounds from model soil was studied using an electrically heated static oven built in-house, using approximately 18 kg of sample in each experiment. Uncontaminated brown soil ("kambizem") from the foothill area of West Bohemia was applied as the model soil. These soils contain a higher proportion of clayey components (>60%). They also have oxides of aluminum and iron and a very small proportion of humic substances (<0.5% wt.). The sample contained less than 35% of fine particles - below 100 µm, and was screened to a particle size below 2 mm. The sample contained 5.1% of free (hydro) oxide iron expressed as Fe₂O₃ and 7.2% of aluminosilicates expressed as aluminum oxide.

Prior to an experiment, a calculated amount of model contaminants (xylene, 2-methylnaphthalene or motor oil) and 1 wt. % of oxidant (benzoyl peroxide) were mixed with model soil. After that, the soil was heated up to 380 °C (temperature gradient 6 °C/min) under air flow (0.6 m³/h) whilst temperature in the oven and in the soil were monitored by thermocouples. Alternatively, samples of soil were spiked with individual contaminants and treated according to the above mentioned procedure.

BTX: The soil was spiked with xylene (Sigma–Aldrich), initial nominal concentration was 10 g/kg of the soil, benzoyl peroxide was added in the amount of 1% weight relative to the soil.

PAH: Initial nominal concentration of 2-methyl naphthalene (Sigma–Aldrich) was 2 g/kg of the soil, concentration of benzoyl peroxide was 1% weight relative to the soil.

NEL: 10 g of synthetic oil (Castrol edge professional BMW LL04 0W-30, Fluid Strength Technology) per 1 kg of soil and hydrogen benzoyl peroxide were added to the soil (1% of weight relative to the soil).

More details are shown elsewhere (Solcova et al., 2014).

Before and after the experiment, soil samples were taken for analyses and the exhaust vapours were condensed and analysed for contaminants. Liquid condensate was subsequently tested in biodegradation experiments employing *Pseudomonas aeruginosa* as a biodegradation agent. Serum bottles (100 ml) containing 20 ml of mineral salts medium (MSM) consisting of essential macro- and micro-nutrients (0.4% NH₄NO₃, 0.47% KH₂PO₄, 0.0119% Na₂HPO₄, 0.001% CaCl₂·2H₂O, 0.1% MgSO₄·7H₂O, 0.001% MnSO₄·4H₂O, and 0.0015 FeSO₄·4H₂O) at pH 7.0 with phosphate buffer supplemented with 10 ppm of YE were used for the experiments. Condensates captured after thermal desorption of contaminated soils were inoculated with *Pseudomonas* sp. strain to a final OD 600 of 0.025 (Kastanek and Demnerova, 1995; Demnerova et al., 1997).

Bioluminescence inhibition using *Vibrio fischeri* as a method for determining toxicity of degradation products was applied (Vosahlikova et al., 2006). The test of bioluminescence inhibition has been standardized for the determination of bioluminescence inhibition of *Vibrio fischeri* bacteria strain. Toxic compounds in solutions cause an inhibition of luminescence. For this purpose the gram-negative bacteria strain *V. fischeri* NRLL-B-11177 is commonly used as a testing microorganism.

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