Chemosphere 114 (2014) 84–92

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Polychlorinated biphenyls removal from contaminated soils using a transportable indirect thermal dryer unit: Implications for emissions

Bing Yang [a], Nandong Xue [a,*], Qiong Ding [b], Rolf David Vogt [c], Lingli Zhou [a], Fasheng Li [a], Guanglong Wu [b], Shilei Zhang [a], Dandan Zhou [d], Bo Liu [a], Yunzhong Yan [a]

[a] State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012, China
[b] Foreign Economic Cooperation Office, Ministry of Environmental Protection, Beijing 100035, China
[c] Department of Chemistry, University of Oslo, P.O. Box 1033, Blindern, Oslo 0315, Norway
[d] Taizhou Academy of Environmental Science Design Research, Taizhou 318000, China

HIGHLIGHTS

- We address in situ PCBs removal from soils using a transportable thermal dryer.
- Concentrations of soil PCBs are reduced by 3 orders of magnitude to 0.08–0.15 $\mu g\ g^{-1}$.
- The emissions to the atmosphere from the unit are below current PCBs regulations.
- The unit is suitable for cleaning regionally dispersed sites of PCBs contamination.

ARTICLE INFO

Article history:
Received 25 August 2013
Received in revised form 13 March 2014
Accepted 30 March 2014

Handling Editor: H. Fiedler

Keywords:
Polychlorinated biphenyls (PCBs)
Soil remediation
Removal efficiency
Thermal desorption
Emission

ABSTRACT

An assessment in China of the application of a transportable indirect thermal dryer unit for the remediation of soils contaminated with polychlorinated biphenyls (PCBs) demonstrated that it is well suited to remove PCBs from soils. A remarkable reduction of total PCBs in soils from 163–770 $\mu g\ g^{-1}$ to 0.08–0.15 $\mu g\ g^{-1}$ was achieved. This represented removal efficiencies of greater than 99.9% and an approximate 100% removal of the toxic equivalent of the PCBs. Furthermore, the emissions to the atmosphere from the unit were in compliance with current PCBs regulations. In conclusion, remediation of PCBs-contaminated soils based on a transportable indirect thermal dryer unit appears to be a highly efficient and environmentally sound treatment technology that has huge implications for cleaning thousands of regionally dispersed sites of PCBs contamination in China.

1. Introduction

Polychlorinated biphenyls (PCBs) are persistent organic pollutants (POPs) and exist as complex mixtures in the environment with up to 209 congeners (Hornbuckle and Robertson, 2010). Taking advantage of their thermal and chemical stability PCBs were used in transformers, capacitors and electric motors. In recent decades, PCBs in the environment have raised a concern because of their persistence, and more importantly, potential adverse toxic effects on bacteria (Abraham et al., 2005), fungi (Sietmann et al., 2006), exoenzymes (Takagi et al., 2007), wildlife (Sierra et al., 2003) and humans (Park et al., 2007; Bergkvist et al., 2012). PCBs are today found ubiquitously in the environment although the use of PCBs was banned internationally through the Stockholm Convention on POPs in 2001.

Extremely high concentrations of PCBs in contaminated hotspots have been found in soils worldwide as a result of industrial activities (e.g. Liu and Liu, 2009; Hornbuckle and Robertson, 2010; Tehrani and Van Aken, 2013). Electrical- and electronic equipment waste (e-waste), being recycled in massive quantities, is considered to be a major cause for heavily PCB polluted soils in China and other developing countries, such as India, Philippines, and Pakistan (BAN and SVTC, 2002; Xing et al., 2011). Zhang et al. (2009) found that total PCBs concentrations in soils from a major site for recycling transformers on the Yangtze River Delta are in

* Corresponding author. Tel.: +86 10 84912638; fax: +86 10 84915187.
E-mail address: ndxue2013@gmail.com (N. Xue).

http://dx.doi.org/10.1016/j.chemosphere.2014.03.131
0045-6535/

the range of 11–153 ng g^{-1}. Similar high contaminations are found in Guiyu (Leung et al., 2006), Taizhou (Shen et al., 2009), and Qingyuan (Liu et al., 2013).

In addition to e-waste recycling sites, burial sites containing capacitors are another cause for soils that are heavily polluted with PCBs by seepage. Investigations of such sites located in the provinces of Zhejiang and Gansu showed mean PCBs concentrations in soils of roughly 2344 and 4983 μg g^{-1}, respectively (Liu et al., 2013; Yang et al., 2013). Thousands of such capacitor-burial sites are regionally dispersed in China, contributing a large potential environmental risk (Yang et al., 2013). Furthermore, this risk is expected to increase as these burial sites are reaching or exceeding their pre-designed working life (⩽20 yrs). As a result, the number of sites with soils contaminated with PCBs that needs to be treated is large and will continue to increase. Growing environmental awareness, driven by the extreme health risks that PCBs impose, has led to an increase in the need to establish a safe and practical soil remediation method (Norris et al., 1999; USEPA, 2012).

Currently, several incineration (e.g. Ikonomou et al., 2002; Weber et al., 2002) and non-incineration (e.g. Aresta et al., 2003; Lundin and Marklund, 2007) technologies are developed for the destruction of PCBs from waste. However, Zhao et al. (2012) points out that incineration results in chlorination reactions even at lower temperatures, while a limited number of new congeners, possibly as the more toxic dibenzofurans (Thacker et al., 2010), are formed at high temperatures. Moreover, incineration is not an applicable technology to use for remediation of large volumes of PCB contaminated soil due to high cost. Instead, thermal desorption and capture of PCBs from contaminated soils (Ishikawa et al., 2007; Lundin and Marklund, 2007), either by In-Situ Thermal Desorption (ISTD) or its ex-situ version as In-Pile Thermal Desorption (IPTD), have produced promising results (e.g. Iben et al., 1996; Vinegar et al., 1997; Aresta et al., 2008). Aresta et al. (2008) reports of a maximum decontamination from PCBs in landfill soils close to 100% under optimum operation conditions using thermal desorption technology. Thermal desorption technologies are also an affordable and sustainable treatment option for POPs waste including PCB-oils, obsolete pesticides, and soils contaminated with POPs (DDT, aldrin, dieldrin, and pentachlorobenzene) (Vinegar, 1998; UNEP, 2004). However, in many developing countries including China, most thermal desorption units are still at a laboratory scale and are currently not cost-efficient. Significant improvements in these technologies need therefore to be made to allow the efficient remediation of PCBs contaminated sites in a cost-efficient manner. Moreover, there is a need for transportable units that are thereby suitable for application at the large number of small-scale PCB contaminated sites across China.

Responding to these needs the State Environmental Protection Administration of China conducted in 2011 a pilot action of "Cleanup Plan for PCBs Burial Sites" that resulted in the construction of a transportable indirect thermal desorption unit. This unit has a number of features that inherently enhance its applicability for remediation of soils containing PCBs. One of the features is that the desorption takes place at relatively low temperatures (just above 500 °C for several minutes). This is important in order to prevent structural changes and calcination that should render the soil less suitable for use (Ishikawa et al., 2007; Lundin and Marklund, 2007). Another is that desorption occurs in an anoxic environment to avoid the formation of toxic species like dioxin (Risoul et al., 2002; Sato et al., 2010). Good turbulence and mixing conditions ensure more homogeneous and complete processing. Despite the presence of these features, the transportable unit has a major concern when remediating PCBs contaminated soils that could lead to increased emissions of PCBs to the air. However, there are limited data on PCBs emissions to air from remediation facilities based on thermal desorption.

This article attempts to address this gap by a test of thermal desorption of PCB contaminated soils in a transportable facility based on an indirect thermal dryer unit. The purpose of this study was to assess the feasibility and performance of this transportable and thus presumably less costly technology for the remediation of regionally distributed sites with PCBs-contaminated soils. Special attention was given to measuring removal efficiency and emission of PCBs to air. The obtained results are assessed in the perspective of using this technology to facilitate a nationwide toxic substance control act for the remediation of numerous small scale PCBs-contaminated soils sites across China.

2. Materials and methods

2.1. Site description

A generic flow diagram of the transportable plant for thermal desorption of PCBs from contaminated soils is shown in Fig. S1. The plant is constructed on a basis of an indirect thermal dryer (ITD-3) unit (Beaudin Consulting, USA). In 2011 the plant was set up at a PCB contaminated site in Jiande City (119°15′56″ E, 29°27′53″ N), about 120 km southwest of Hangzhou City, the provincial capital of Zhejiang Province. No apparent pollution source was observed nearby the plant. The whole facility covers an area of about 2000 square meters, of which the ITD-3 unit had a length of 12.2 m, width of 2.38 m, and height of 2.91 m. The ITD-3 consists of a conveyor belt transporting the soil contaminated with PCBs into a heated desorption treatment chamber that is set under vacuum and flushed with inert nitrogen gas. From the results of pre-experiments an optimum average temperature of 430 °C (range: 408–502 °C) in the desorption chamber was selected for the thermal desorption tests. The gas from the treatment chamber, containing released PCBs, are exhausted into a washing/cooling system for condensation and capture of the PCBs. After being stripped of PCBs, the off-gas pass through an off-gas treatment system (an activated carbon filter as well as a particle filter) prior to release to the atmosphere. The condensed PCBs are stored in cooled collection tanks and sent for incineration at the Su Giatun experiment base in Shenyang City, constructed by Shenyang Academy of Environmental Sciences in 2011.

The ITD-3 was designed for remediating PCBs contaminated soils (⩽1000 mg kg^{-1}) with a treatment capacity of 72 tons of contaminated soils per day and an annual operation of 300 d. The system design, being robust and simple to ship, is believed to be possible to dismantle and made ready for transport within 3 d.

2.2. Sample collection

Soil sampling along the process line was performed following Chinese National Guidelines for Technical Specification for Soil Environmental Monitoring (HJ/T 166-2006). Gas samples of inlet gas and off-gas, as well as from the local environment, were collected following Chinese National Guidelines for Ambient Air Quality Monitoring (HJ/T 194-2005). Sampling details are summarized in Table 1.

Air samples from the inlet (In) and outlet (Out) of the conveyor belt, as well as the off-gas (Off-gas) were used to assess the emissions of PCBs to the atmosphere from the facility (Fig. S1). In addition, control samples (Con) of air were collected close to the facility (30 m) and at a more distant site (700 m), in the village of Tongxi. Three air sampling campaigns from inlet (In1 to In3) and outlet (Out1 to Out3) of the conveyer belt, six sampling campaigns from off-gas (Off1 to Off6), and two sampling campaigns from control air close to the facility (Con1-1 and Con1-2) and at some distance (Con2-1 and Con2-2), were conducted. Parallels were collected of