



Technical Note

Disposal of historically contaminated soil in the cement industry and the evaluation of environmental performance



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HIGHLIGHTS

- Contaminated soil was co-processed without sacrificing cement clinker quality.
- PCDD/PCDF emissions ranged from 0.0023 to 0.0085 ng I-TEQ Nm⁻³.
- DRE and DE of DDTs/HCHs were better than 99.9999% and 99.99%, respectively.

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ABSTRACT

Approximately 400 000 t of DDTs/HCHs-contaminated soil (CS) needed to be co-processed in a cement kiln with a time limitation of 2 y. A new pre-processing facility with a “drying, grinding and DDTs/HCHs vaporizing” ability was equipped to meet the technical requirements for processing cement raw meal and the environmental standards for stack emissions. And the bottom of the precalciner with high temperatures >1000 °C was chosen as the CS feeding point for co-processing, which has rarely been reported. To assess the environmental performance of CS pre- and co-processing technologies, according to the local regulation, a test burn was performed by independent and accredited institutes systematically for determination of the clinker quality, kiln stack gas emissions and destruction efficiency of the pollutant. The results demonstrated that the clinker was of high quality and not adversely affected by CS co-processing. Stack emissions were all below the limits set by Chinese standards. Particularly, PCDD/PCDF emissions ranged from 0.0023 to 0.0085 ng I-TEQ Nm⁻³. The less toxic OCDD was the peak congener for CS co-processing procedure, while the most toxic congeners (i.e. 2,3,7,8-TeCDD, 1,2,3,7,8-PeCDD and 2,3,4,7,8-PeCDD) remained in a minor proportion. Destruction and removal efficiency (DRE) and destruction efficiency (DE) of the kiln system were better than 99.9999% and 99.99%, respectively, at the highest CS feeding rate during normal production. To guarantee the environmental performance of the system the quarterly stack gas emission was also monitored during the whole period. And all of the results can meet the national standards requirements.

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1. Introduction

In recent years, China has devoted great effort to remediate soil contaminated by industrial enterprises after they have relocated during rapid urbanization. The soil we excavated for treatment was from a closed pesticide manufacturing plant where the main contaminants were DDTs and HCHs. An integrated and systematic sampling and pollutants analysis of the brown land was taken place for environmental impact assessment by the land owner. The land was divided into 45 small squares by 50 m × 50 m, soil

samples from each square have been taken for different height varying from 0 to 1.8 m as the first layer, 1.8–5 m as the second layer and 5–9 m as the third layer. The highest concentration of DDT was 33548.14 mg kg⁻¹ in the first layer and 4661.46 mg kg⁻¹ for HCHs in the second layer. The average concentration for DDT and HCHs was 554.852 mg kg⁻¹ and 23.819 mg kg⁻¹ in the first layer, 139.169 mg kg⁻¹ and 56.07 mg kg⁻¹ in the second layer, and 1.367 mg kg⁻¹ and 0.533 mg kg⁻¹ in the third layer.

Conventional methods for destroying organic pollutants comprise biological, chemical and thermal treatments (Cravotto et al., 2007). Biological and chemical technologies are often time consuming and not cost-effective for treating large volumes of polluted materials (Cravotto et al., 2007; Lai et al., 2009; Zhang

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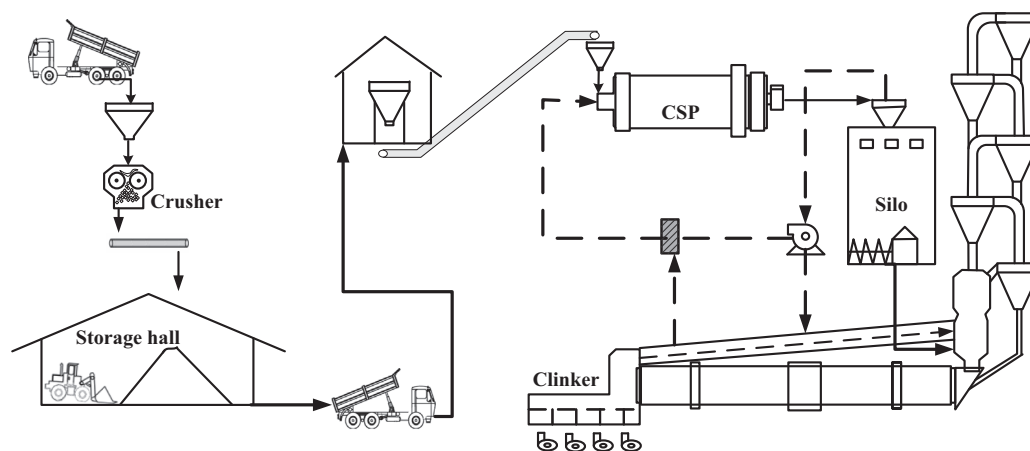


Fig. 1. Flow chart of the CS pre and co-processing unit operations.

et al., 2010; Venny et al., 2012). Thermal decontamination of soil is widely used due to many advantages, including removal efficiencies above 99%, short remediation times and applicability to a wide range of organic contaminants (Lee et al., 1998; Gan et al., 2009; Chien, 2012). In this field-scale remediation case, the survival of the degrading organisms can be affected by the toxicity associated to highly contaminated sites (Rein et al., 2007; Perelo, 2010; Megharaj et al., 2011). The cement kiln is one thermal treatment that has attracted much attention for the environmentally sound destruction of persistent organic pollutants (POPs), owing to its inherent features such as high temperatures, long residence time and surplus oxygen as well as the elimination of slag, ash or liquid residue byproducts (Chadbourne, 1997; Reijnders, 2007; Karstensen, 2008; Kookos et al., 2011). Several documents have demonstrated that this process can simultaneously offer more disposal capacity, maintain good destruction efficiencies and avoid the formation of PCDD/PCDFs (Karstensen et al., 2006, 2010; Weber, 2007; Karstensen, 2008; Khumsaeng et al., 2013).

Because the site was located in the city center, the site cleanup was required to be completed within 2 y. To meet this time limitation, co-processing by cement kiln was chosen as the disposal method for the soil with DDT and HCH concentrations higher than 50 mg kg^{-1} . The total amount was estimated to be as high as 400 000 t. For the soil with DDT and HCH concentration lower than 50 mg kg^{-1} was remedied in-situ by biological method.

The chemical components of DDTs/HCHs-contaminated soil are similar to clay, thus after pre-processing, this soil can be co-processed as an alternative raw material in the cement kiln to destroy DDTs/HCHs and recover valuable SiO_2 components during clinker sintering. Generally, alternative raw materials are fed at the raw mill (GTZ and Holcim, 2006; Karstensen, 2008). In this manner, their fineness, moisture and homogenization with normal raw meal can be controlled to guarantee the clinker quality. However, alternative raw materials containing components that are volatile at low temperatures have to be fed into the high temperature zones of the kiln system. If the soil would have been fed to traditional points at low temperature, such as the raw mill and the raw meal feeding point at the outlet of preheater, DDTs/HCHs would not have decomposed immediately. Instead there would have been a high risk of incomplete destruction or escape of DDTs/HCHs to the stack. Therefore, a higher temperature location, the bottom of the precalciner ($>1000 \text{ }^\circ\text{C}$) (Li et al., 2009), was modified and chosen as the feeding point. This feeding point enables instant homogenization with raw meal and can assure DDTs/HCHs destruction efficiency and clinker quality. So far, feeding CS into the precalciner of a cement kiln has rarely been reported in the literature (Yan et al., 2014).

CS cannot be used directly as an alternative material, as it must first undergo a preparation process. This step produces a waste product with defined characteristics that complies with the technical specifications of cement production and guarantees that environmental standards are met (GTZ and Holcim, 2006). Due to the large particle size and high moisture content of virgin CS, grinding and drying procedures are required in a specialized treatment facility with high pre-processing capacity. To prevent the output of DDTs/HCHs during the drying procedure, the thermal desorption system, “drying, grinding and DDTs/HCHs vaporizing” facility (Li et al., 2011), was designed and employed to fulfill the task.

Based on the above analysis and previous experience that precalciner are known to be efficient and environmentally sound feeding points for complex organic pollutants (GTZ and Holcim, 2006; Karstensen et al., 2006), the precalciner of the cement plant was chosen as the feed point and particularly modified to serve this task. A CS co-processing system with 35 t h^{-1} pretreatment capacity was constructed, including a CS pre-processor (CSP) with drying/grinding ability and an efficient, environmentally sound precalciner was equipped for co-processing (Li et al., 2012a). After running for more than 1 y, 218 078 t of CS have been co-processed. In this period, a 2 d test burn and regularly kiln stack gas emission monitoring were implemented. Third parties monitored the clinker quality, CSP effectiveness, destruction and removal efficiency (DRE), destruction efficiency (DE) for DDTs and HCHs and stack emissions (including organic compounds, acid gases and particulates) to assess the performance of the system during the test burn. Quarterly kiln stack gas has been sampled and analyzed for DDTs, HCHs, TVOC and dust emission. A baseline of DDTs, HCHs, PCDD/PCDFs emission was measured on October 30, 2011 before CS disposal.

2. Materials and methodology

2.1. CS pre and co-processing procedure

As shown in Fig. 1, first, CS is crushed to a size less than 100 mm and stored in a pre-homogenization hall. Second, the crushed soil is dosed and fed into the CSP, where the soil is ground to be sifted through a $200 \mu\text{m}$ sieve and dried using $300\text{--}350 \text{ }^\circ\text{C}$ air from the tertiary air duct and circulating fan. Because the boiling points of DDTs and HCHs range from 260 to $330 \text{ }^\circ\text{C}$, the hot air in the CSP could partially vaporize the contaminants. These vaporized components are immediately taken to the precalciner and destroyed. Thus, grinding, drying and removing a part of the organic pollutants are realized in parallel. The pretreatment process could be

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