Test burn with PCB–oil in a local cement kiln in Sri Lanka

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ABSTRACT

The production and use of polychlorinated biphenyls (PCBs) have ceased and most developed countries have disposed off their stocks long time ago. PCBs can however still be found in the environment and one important source is accumulated stocks in developing countries. Sound treatment of PCB is costly and most developing countries do not have dedicated hazardous waste incinerators or non-combustion technologies available for domestic disposal and can usually not afford export.

High temperature cement kilns have been used to treat organic hazardous wastes in developed countries for decades and shown to constitute a sound option if well managed and controlled. In contrast to dedicated hazardous waste incinerators and other treatment techniques, cement kilns are already in place in virtually every country and may constitute a treatment option. The objective of this study was therefore to carry out the first test burn with PCB–oil in a developing country cement kiln and to assess its feasibility and destruction performance.

The 3 d test burn demonstrated that the Sri Lankan cement kiln was able to destroy PCB in an irreversible and environmental sound manner without causing any new formation of PCDD/PCDF or HCB. The destruction and removal efficiency (DRE) was better than 99.9999% at the highest PCB feeding rate.

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1. Introduction

Commercial production of PCB started in the US, Germany and France in the 1930s and ceased when the production in Russia ended in 1993 (de Voogt and Brinkman, 1989; AMAP, 2000). The total volumes produced are uncertain; estimates vary from 60 000 tons to about 2 million tons (Fiedler, 2001; Holoubek, 2001; Breivik et al., 2002a). PCB production was banned and phased out in the US and European countries during the 1970s and it can be anticipated that most developed countries now have disposed off their stocks, primarily by using thermal techniques (Rosiers, 1983; Kokoszka and Kuntz, 1985; Carpenter and Wilson, 1988).

Most developing countries do not have dedicated hazardous waste incinerators or non-combustion technologies available for treatment of hazardous chemicals like PCBs and can usually not afford export due to high costs; e.g. Thailand exported 760 tons of PCB to Europe for high temperature incineration at a cost of 5000 US$/ton in the period 1992–2002; still approximately 150 tons remains in the country (Chotchamlong, 2008).

If not stored properly, these untreated stocks represent a constant influx of PCBs to the global environment (Breivik et al., 2002b, 2004; Van der Gon et al., 2007). Several international conventions aiming to protect human health and the environment, acknowledge that developing countries generally need to strengthen their national capabilities and capacity on sound management of hazardous chemicals like PCBs (UNECE, 1998; UNEP, 2004). The intention of the Basel Convention, ratified in 1989, is to control shipment of hazardous wastes across borders, to avoid dumping in developing countries and to stimulate local treatment.

The Basel Convention (2007) has described 12 technologies (including high temperature incinerators and cement kilns) assumed to be feasible for environmentally sound treatment of PCBs. The techniques can broadly be divided into two main groups, thermal and chemical treatment (non-combustion); a comparison of the technologies is however lacking making it difficult to compare the performance and feasibility.

Non-combustion technologies seem to be gaining popularity compared to thermal treatment and relatively large sums are allocated to fund non-combustion technologies in developing countries despite limited experience (GEF, 2008). Super critical water oxidation (SCWO) is for example referred to by UNEP (2004) as a "commercialised technology with considerable experience"

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despite the fact that a full-scale 100 million US$ SCWO plant had to close in the US due to high operation costs, corrosion problems and high releases of PCDD/PCDFs (Weber, 2007). Problems with PCDF formation and inferior destruction efficiency have been experienced in other SCWO studies (Hatakeda et al., 1999; Weber, 2004a,b; Seok et al., 2005; Kawasaki et al., 2006). A recent article states that the operating conditions for a number of non-combustion technologies have the potential to generate high concentrations of PCDD/PCDF, for example when treating PCBs (Weber, 2007).

Decades of experience with thermal treatment techniques, like incinerators and cement kilns, have on the other side taught us how to simultaneously achieve good destruction efficiencies and to avoid the formation of PCDD/PCDFs (Ahling and Lindskog, 1978; Kokoszka and Kuntz, 1985; Dickson et al., 1989; Kilgroe et al., 1990; Hagenmaier, 1991; Addink and Olie, 1995; Buekens and Huang, 1998; Hunsinger et al., 2002, 2003; McKay, 2002; Weber, 2007). A stringent regulatory framework is also in place to monitor and control the performance; e.g. the US Toxic Substances Control Act (TSCA) PCB incineration criteria require a temperature of 1200 °C and 2 s retention time at 3% oxygen for thermal treatment, as well as the completion of a test burn demonstrating a destruction and removal efficiency (DRE) of 99.9999% for PCBs (Federal Register, 1999; Lee et al., 2000).

Cement kilns possess many inherent features which makes them suited for organic hazardous chemicals treatment; such as high temperatures, long residence time (>1200 °C for several seconds), surplus oxygen during and after combustion, good turbulence and mixing conditions, no generation of by-products such as slag, ashes or liquid residues and complete recovery of energy and raw material components in the waste (Chadbourne, 1997; Karstensen et al., 2006). Test burns with PCB in cement kilns were conducted in Europe and North America in the 1970s and have normally shown compliance with the US TSCA requirements (Ahling, 1979; Black and Swanson, 1983; Viken and Waage, 1983; Benestad, 1989; Karstensen, 1998). However, many cement plants have chosen not to treat PCB since the 1980s due to controversy and fear of bad publicity (Karstensen, 2008). In fact, newer results of test burns with PCBs using thermal treatment techniques, like incinerators and cement kilns, cannot be found in the literature.

Despite the obvious need, no studies have up to now investigated the feasibility of PCB-destruction using cement kilns in developing countries. In contrast to incinerators and other treatment techniques, cement kilns already exists in virtually every country and may represent a powerful treatment potential for organic hazardous chemicals (Karstensen et al., 2006). The objective of this study was therefore to carry out a test burn with PCB–oil in a local cement kiln under real developing country conditions and to assess its feasibility and destruction performance.

2. Materials and methods

The bulk of Sri Lanka's PCB came from emptying of transformers belonging to the state owned Ceylon Electricity Board; drums with PCB–oil had been stored in a warehouse in Colombo for more than 20 years waiting for a disposal solution to emerge.

2.1. Preparation of a PCB-diesel mix for the test burn

High concentration PCB–oil was kept in 60 L stainless steel drums; diesel washings from cleaning of the transformers, in 200 L steel drums. The PCB–oil was confirmed to be Pyralene with an average concentration of 59% of PCB, 36% trichlorobenzene and 5% tetrachlorobenzene.

Cement kilns usually have limited tolerance for chlorine and the total input needs to be controlled to avoid process clogging and impacts on the product quality. A blend of the high concentration PCB–oil and the diesel-washings was prepared at the storage site and transported in two chemical resistant tanks to the cement plant located in Puttalam, 120 km North West of Colombo.
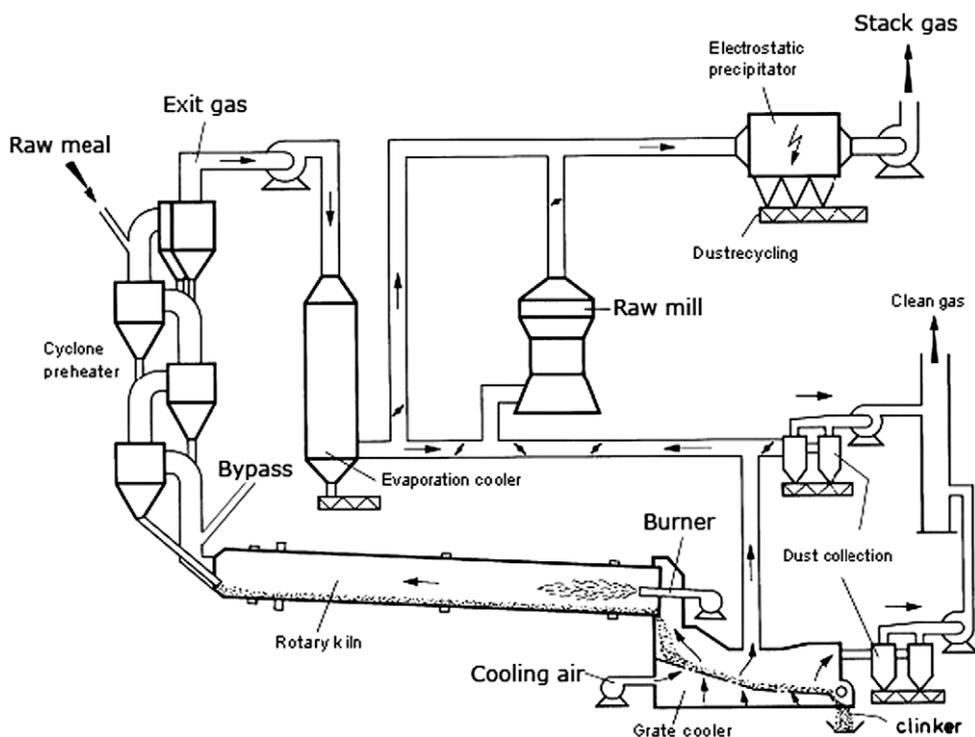

Fig. 1. Puttalam cement kiln with cyclone preheater, bypass and gas dust collection.

Fig. 2. Gas and material temperature of the individual stages of a cyclone preheater.

The mix of PCB–oil and diesel-washings from Colombo was further blended to a total 10 000 L with diesel oil in a steel feeding tank at the cement plant; the final PCB-diesel oil mix was homogenised and fed directly to the main burner flame of the cement kiln during two consecutive days of testing with various feeding rates and PCB-concentration. The test burn started with emission measurements when no PCB was fed to the kiln, followed by Test day 1 with a feed rate of $500 \, L \, h^{-1}$ of PCB-diesel oil mix with 14 000 mg PCB L^{-1} and Test day 2 with a feed rate of $1000 \, L \, h^{-1}$ of PCB-diesel oil mix with 10 050 mg PCB L^{-1}. The feeding system was calibrated and tested before start up.

2.2. Cement plant in Sri Lanka

Holcim Puttalam is the only production plant in Sri Lanka that produces cement clinker. The plant has two dry-process kilns, both equipped with a four-stage 54 m high single string suspension pre-heating tower. The main fuel is coal, but normally 15% of the heat requirement is substituted with the alternative fuels derived from wastes. The plant is equipped with state-of-the-art process control equipment, the latest technology in air pollution control (electro static precipitators (ESP) and bag-house filters) and on-line emission monitoring\ of gross air pollutants. All dust collected in the air pollution control devices are recovered in the process; i.e. the plant does not produce any residues which need to be disposed off.

The rotary kilns are 54 m long, 3.5 m in diameter and have a production capacity of 850 tons clinker per day. From the top of the cyclone preheating tower, the finely crushed alkaline raw meal powder is fed counter currently with the hot kiln exhaust gases, i.e. the preheating tower is practically acting as a large dry lime scrubber. The limestone is completely calcined as it moves down the kiln towards the burning zone were the material melts to form clinker

Fig. 3. Temperatures and gas residence time in a preheater kiln; gas temperature is underlined.

at a temperature of 1450 °C. The clinker is cooled by air in the clinker cooler and the hot air is recovered in the process. The exit gas from the kiln and the preheater is normally directed through the raw mill and the coal mill for drying purpose.

2.3. Feeding control and safeguards

The technical conditions and the process chemistry had been evaluated prior to the test and considered to be feasible for carrying out a test burn with PCB-diesel oil mix; the receiving, handling, storage and feeding of PCB-diesel oil mix, as well as the power and water supply, were considered to be stable, safe and adequate. The following preconditions had been agreed prior to the test:

- The PCB-diesel oil mix should be fed through the main burner flame (not during kiln start-up, shutdowns or major upsets).
- The PCB-diesel oil mix should be thoroughly homogenised prior to feeding and the heating value compensated for by reducing the coal feed.
- The process should be run in compound mode operation, i.e. the exit gas directed through the raw mill and under identical process conditions all 3 d.
- The coal-mill should be off and the bypass should be in stable operation.
- Alternative fuels and raw materials should not be used.
- All involved personnel should have been trained prior to the test.

Process controls were implemented and should provide for automatic shutdown of the PCB feed if any of the following conditions occurred:

- Interruptions of the coal feeding.
- Interruptions in combustion air flows.
- Kiln inlet temperature below 900 °C and outlet temperatures below 1200 °C.
- Positive pressure in the burning zone of the kiln.
- Kiln inlet oxygen content less than 2.5%.
- Refractory damage, ring formation in the kiln or cyclone blockage.

2.4. Test burn outline and process conditions

The test was carried out in kiln # 2 over 3 d, 2–4 August 2006 and should compare the emissions under Baseline conditions, i.e. when feeding coal only, with the emissions when coal and a PCB-diesel oil mix were fed at a rate of 500 and 1000 L h^{-1}, amounting to 7 and 10.05 kg PCB per hour for the two test days respectively (corrected for density). The entire test burn was inspected by university experts, central and provincial authorities, Holcim personal and third party experts.

2.5. Sampling and monitoring

The feeding of the PCB-diesel oil mix to the kiln started 4 h before the stack gas sampling started to ensure stable conditions. The exit gases were sampled (>5 h) simultaneously from the main stack and the bypass (except the third day) by an accredited testing company from Australia. The USA EPA method 23 was used for PCDD/PCDFs and the USA EPA method SW-846 for PAHs, PCBs and HCB (Federal Register, 2000; Abad et al., 2006). Emissions were continuously monitored for O_2, CO and NO_x in the kiln inlet and the preheater outlet gas; and for O_2, CO, CO_2, NO, NO_2, SO_2, HCl, NH_3, H_2O and VOC in the stack exit gas.

3. Results and discussion

A summary of the main stack results of the test burn is given in Table 1.

3.1. Destruction of PCB

The PCB-diesel oil mix was fed through the main burner flame, i.e. being exposed to a gas phase temperature of approximately 2000 °C. Already in the 1960s Lidget and Vodden (1970) showed that liquid PCBs were destroyed by incineration at a temperature of 1000 °C and a residence time of 3 s. Ahling and Lindskog (1978) showed that the PCB Pyralene and Arochlor were destroyed within the temperature range of 675–1000 °C provided surplus oxygen and a residence time longer than 2 s.

The DRE of this test was calculated on the basis of mass of PCB fed to the kiln minus the mass of the remaining PCB in the stack emissions according to the following equation:

$$DRE = [(W_{in} - W_{out})/W_{in}] \times 100$$

where W_{in} is the mass of sum-PCB entering the kiln and W_{out} is the sum-PCB exiting the stack gas. The sum-PCB measured in the exit stack varied between 0.09 and 0.178 µg N m^{-3} representing a DRE 99.999944% for Test day 2, when the PCB feeding rate was highest (or >99.999999% when corrected for the background emission). The PCB emissions on Test day 2 were almost identical with the Baseline emissions, when no PCB was fed. The DRE for Test day 1 was slightly lower, 99.999837%, due to a combination of higher stack gas concentration and lower feeding rate. The dioxin-like PCBs varied between 0.0046 and 0.0069 ng N m^{-3} in the stack gas; dioxin-like PCBs were however not measured in the PCB-diesel oil mix.

Solid samples from the production process, i.e. cement clinker and dust from the ESP, were analysed for PCBs; the results were however not conclusive and could not be used for calculating the more comprehensive destruction efficiency (DE).

3.2. Formation of PCB

The fact that PCBs were found in the exit gas at same concentration when no PCB was fed to the kiln as when feeding 10.05 kg PCB h^{-1}, indicates that PCBs are formed in the process. PCB emissions are normally not regulated for cement kilns and the availability of emission data in the public domain is very limited. Measurements of 13 German cement kilns in 2001 showed normal baseline concentrations up to 0.4 µg of sum-PCB N m^{-3}; twice as much as found in this study (VDZ, 2002). The concentration of dioxin-like PCBs found in this study was at the same concentration level identified in normal cement kiln emissions by other researchers (Alcock et al., 1998, 1999; Luthardt et al., 2002); see also Ahlborg and Hanberg (1994).

It has previously been shown that organic compounds present in the raw materials (e.g., clay, shale, gypsum, etc.) are the major

Table 1
Main stack results corrected to 10% O_2 and standard conditions.

	Baseline	Test 1	Test 2
Fuel and PCB-feed:			
– Coal (tons h^{-1})	5	4.3	3.5
– PCB-diesel oil mix (L h^{-1})	0	500	1000
– Sum PCB (kg h^{-1})	0	7	10.05
Exhaust gas volume (Nm^3 h^{-1})	56 000	64 000	62 000
Sum PCBs (µg N m^{-3})	0.090	0.178	0.091
Dioxin-like PCBs (ng TEQ N m^{-3})	0.0046	0.0069	0.0051
PCDD/PCDF (ng I-TEQ N m^{-3})	0.018	0.016	0.0087
Sum chlorobenzenes (µg N m^{-3})	2.6	2.7	2.5

sources of organic emissions from cement kilns. These compounds, primarily hydrocarbons, are volatilised from the raw materials in the preheater region prior to entering into the high temperature kiln (Trenholm and Hlustick, 1990; Dellinger et al., 1993; Schreiber and Strubberg, 1994). Chlorination of these organic compounds into chlorobenzenes and aromatic precursors may be the primary source of PCB and PCDD/PCDF (Sidhu and Dellinger, 1995).

Lemieux et al. (2001) and others have shown that PCDD/PCDF and PCB are formed by more or less the same formation mechanisms in combustion systems (Addink and Olie, 1995; Luthardt et al., 2002; Liu et al., 2004; UNEP, 2005; Abad et al., 2006; Pandelova et al., 2006; Ishikawa et al., 2007). The detailed understanding of PCDD/PCDF and PCB formation mechanism in cement production is not yet complete, but it seems that a combination of heterogeneous surface catalysed reactions in the preheater and the post-preheater zones are the most important; de novo synthesis is probably of less importance (Dickson et al., 1989; Karstensen, 2008).

A preheater can be viewed as chemical reactor were finely ground raw materials, with adsorbed and absorbed organic compounds, will move down the preheater towards higher temperatures, leading to a volatilisation of the organic compounds in the raw materials in the lower part of the preheater. The organic compounds will then be brought back to the top of the preheater with the exit gas from the kiln, which moves counter currently to the raw material flow, and adsorb and absorb on the introduced raw material and dust particles at the lower temperature at the top of the preheater, creating a complex sorption–desorption circulation process. The net formation of PCDD/PCDFs and PCBs is therefore probably due to a complex combination of simultaneous formation and decomposition reactions were:

- PCBs and PCDD/PCDFs are destroyed in the high temperature zone of the kiln region.
- Organic compounds in the raw materials are volatilised when introduced into the preheater leading to the formation of products of incomplete combustion (PICs) and chlorinated organics in the preheater, shown by the formation of chlorobenzenes (see Table 2).
- Gas-phase formation reactions between precursors in the lower part of the preheater can form PCBs and PCDD/PCDFs directly.
- Heterogeneous surface-catalysed reactions can form precursors, PCBs and PCDD/PCDFs in the preheater.
- An sorption–desorption circulation process of naturally and adsorbed PCBs and PCDD/PCDFs in the preheater and the raw mill (see Fig. 1).
- Interaction with equilibrium reactions and circulation of chlorine, sulphur and alkali materials in the upper parts of the kiln zone and lower parts of the preheater may lead to inhibition of formation of PCBs and PCDD/PCDFs in the preheater.
- Lastly, de novo synthesis and formation of PCB and PCDD/PCDF in the electro static precipitator and the post-preheater zone; probably at negligible levels (see Figs. 2 and 3).

Table 2
Chlorobenzenes in main stack during 3 d of testing (ng N m^{-3}).

	Baseline	Test 1	Test 2
MCB	NA	NA	NA
1,3-DCB	827	811	719
1,4-DCB	536	486	411
1,2-DCB	469	541	437
1,3,5-TCB	107	92	80
1,2,4-TCB	447	514	462
1,2,3-TCB	123	173	164
1,2,3,4-TeCB	29	30	28
1,2,3,5/1,2,4,5-TeCB	63	62	239
PeCB	14	23	23

3.3. Emission of chlorinated organic compounds

The test burn showed that the PCB–oil had been destroyed without any new formation of PCDD/PCDF or HCB; an implicit requirement of the Stockholm Convention saying that disposal of POPs needs to be "irreversible", i.e. no new POPs should be formed during the destruction (UNEP, 2001). The lowest PCDD/PCDF concentration in the main stack was found under Test 2, when the PCB feed rate was highest; the highest concentration was found under Baseline conditions, when no PCB was fed. None of the measurements exceeded the EU emission limit value of 0.1 ng I-TEQ m^{-3}, in accordance with a recent study evaluating more than 2000 PCDD/PCDF cement kiln measurements and indicating that most modern cement kilns co-processing waste (also organic hazardous wastes) can meet an emission level of 0.1 ng PCDD/PCDF I-TEQ m^{-3} (Karstensen, 2006, 2008).

Chlorobenzenes, except hexa, were identified in the main stack. Table 2 shows that di-chlorinated and tri-chlorinated benzenes dominate; monochlorobenzene was not measured. The Baseline emissions are at the same level as when feeding PCB, a feed that actually contained percentage level of tri- and tetrachlorobenzene. The fact that chlorobenzenes could not be found in the bypass, indicates that chlorobenzenes in the PCB-mix were destroyed in the main flame, but formed again in trace amounts in the lower part of the preheater by chlorination of organic compounds present in the raw material as experienced by others (Lamb et al., 1994; Schreiber and Strubberg, 1994; Sidhu et al., 2001).

Ahling and Lindskog (1978) showed that HCB was formed during PCB-incineration at temperatures approaching 1000 °C and residence time shorter than 2 s, maybe due to the increase of reactive free chlorine radicals; no HCB could however be detected in the emissions of this test burn. HCB has been found in cement kiln emissions, usually below 4 ng HCB m^{-3} (Karstensen, 2006); normal emission range from coal fired power plants is 11.5–42 ng HCB m^{-3} (Grochowalski and Konieczyński, 2008).

Fig. 4 presents the sum of the toxicity equivalents for PCDD/PCDF and PCB and shows that dioxin-like PCBs contribute to the total toxicity in the range of 20–37% in the main stack compared to PCDD/PCDF. Alcock et al. (1998, 1999) found that the PCB TEQ contribution from a cement kiln in the UK, which did testing with substitute liquid fuel, constituted around 38–60% of the total TEQ; the rest was from PCDD/PCDF. Luthardt et al. (2002) found that the dioxin-like PCB contribution from normal cement kiln emissions constituted approximately 14% of the total TEQ compared with PCDD/PCDF.

The major contribution to the total TEQ from PCDD/PCDF in this study came from the lower chlorinated congeners, especially 2,3,7,8-tetrachlorodibenzo-p-dioxin and 2,3,4,7,8-pentachloro-dibenzofuran; consistent with findings of Abad et al. (2004). Among the dioxin-like PCBs, mainly PCB 126 and 169 was detected in the flue gas in this study; the same congeners identified in normal cement kiln emissions by others (Alcock et al., 1998, 1999; Luthardt et al., 2002).

3.4. Bypass stack emissions versus main stack

The concentration of most compounds was below or close to the detection limit in the bypass stack. Low emissions in the bypass stack has been confirmed in several other studies and can be attributed to the fact that the bypass duct draws a small amount of the exit gas (<5% of the total volume) directly from the high temperature kiln (<1000 °C); i.e. survival organic compounds fed through the main burner is unlikely. The exit gases of the main stack on the other side have passed both the preheater and the post-preheater zones and will absorb/adsorb components in the finely
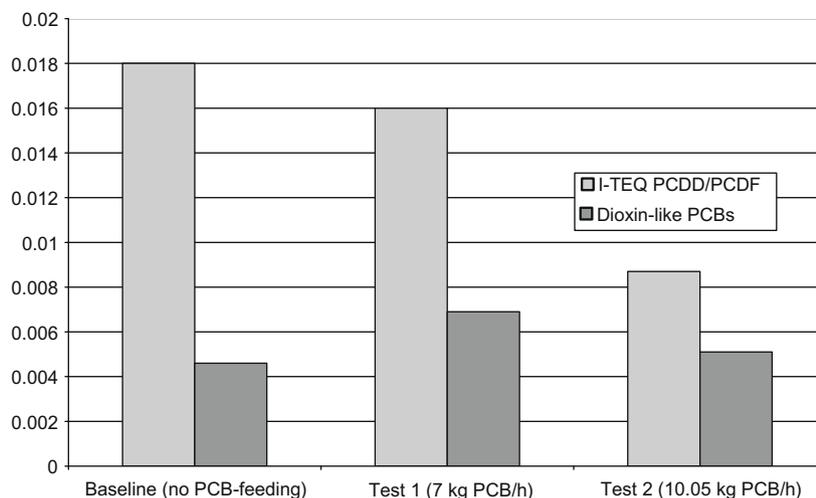
Fig. 4. Sum of I-TEQ PCDD/PCDF and dioxin-like PCBs (WHO-TEQ) in the main stack emissions during the three testing days (ng N m^{-3}).

ground raw meal and therefore have a different composition (Lamb et al., 1994; Schreiber and Strubberg, 1994; Sidhu et al., 2001).

3.5. Emission of other air pollutants

The sum of PAHs was below 0.54 ng N m^{-3} in all samples; benzene could not be detected in any sample (<1 mg N m^{-3}) and the highest concentration of VOC was found in the main stack under Baseline measurement (23 mg N m^{-3}); the concentration was 14 mg N m^{-3} when feeding PCB. Hydrogen chloride was <20 mg N m^{-3} all 3 d; no correlation between the feeding of PCB-diesel oil mix and the emissions of HCl could be established. The ammonia concentration was <0.2 mg N m^{-3} in all samples; carbon monoxide and SO$_2$ below 15 and 5 mg N m^{-3}, respectively all 3 d.

Process instability during the test caused some visual dust emissions from the bypass a few times (not quantified) but the dust levels measured in the main stack were in compliance with Sri Lankan regulation, i.e. below 50 mg N m^{-3}. NO$_x$ was measured at an average concentration of 600 mg N m^{-3}; the on-line monitoring did however not function properly. The concentration of antimony, arsenic, cadmium, chromium, cobalt, copper, lead, manganese, mercury, nickel, thallium, tin, vanadium and zinc measured in the dust and gas phase were at low concentrations, usually below the limit of detection (μg N m^{-3}). Solid process samples, i.e. raw meal, coal, clinker, ESP dust, bag house dust and bypass dust were collected every second hour; chemical analysis and quality testing of the cement and concrete produced during the test showed results within normal ranges; no influence of the PCB-feeding could be identified.

4. Conclusion

No studies have up to now investigated the PCB destruction potential using cement kilns in developing countries. In contrast to incinerators and other treatment techniques, cement kilns already exists in virtually every country and may be found feasible and cost-efficient to treat organic hazardous chemicals and other wastes. Waste treatment in cement kilns implies recovery of energy and valuable raw material components in the wastes, i.e. reducing the need for fossil fuel and virgin raw materials, reducing the overall CO$_2$ emissions compared to land filling, export or to building new treatment options.

The test burn demonstrated that the cement kiln was able to destroy PCB–oil effectively in an irreversible and environmental sound manner, i.e. without causing any new formation and emission of PCDD/PCDFs or HCB. The DRE under the worst PCB feeding conditions was >99.9999% and the overall environmental performance were in compliance with Sri Lankan regulation and international practice.

Acknowledgement

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