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Polychlorinated biphenyl concentration changes in sewage sludge and organic municipal waste mixtures during composting and anaerobic digestion

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HIGHLIGHTS

- The PCBs degradation under anaerobic and aerobic conditions was investigated.
- During composting the loss of PCBs depended on their chlorination level.
- During anaerobic digestion the PCBs loss did not depend on the chlorination level.
- The PCBs degradation without PCB 28 was described by a first-order kinetic model.
- The second-order kinetic model described the loss of the PCB 28 contents.

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ABSTRACT

We determined the changes in polychlorinated biphenyl (PCB) concentrations in a mixture of sewage sludge and the organic fraction of municipal waste during composting and during anaerobic digestion. The processes were carried out on a laboratory scale. The PCBs were analyzed in the waste samples using gas chromatography–mass spectrometry. We evaluated the rates at which the PCB concentrations decreased during composting and during anaerobic digestion and compared the PCB degradation kinetics during these processes. The most important conclusion of this work is that anaerobic digestion is much more effective than composting at removing PCBs from a mixture of sewage sludge and the organic fraction of municipal waste.

1. Introduction

Intensive industrial and agricultural development causes the concentrations of organic pollutants (e.g. polychlorinated biphenyls–PCBs) in biowaste to increase. PCBs are found in the natural environment because of human activities, natural sources of PCBs not having been found (Borja et al., 2005). Approximately 30% (i.e., 10 million tonnes) of the total worldwide production of PCBs has entered the environment (Benabdallah El-Hadj et al., 2007). The presence of these compounds in sewage sludge significantly decreases the potential for the sludge to be used in agriculture. According to proposed changes to EU Directive 1986/278/EEC, the polychlorinated biphenyl (PCB) concentration (expressed as the sum of the concentrations of the indicator congeners, PCBs 28, 52, 101, 118, 138, 153, and 180) should not exceed 0.8 mg/kg dry matter (DM) in sewage sludge used as a crop fertilizer (CEC). Blanchard et al. (2004) performed a study in Paris, France, and found that the sum of the seven indicator PCB concentrations in the sewage sludge from that area did not exceed the approved limit, and that the mean concentration was 0.62 mg/kg DM. Similar results have been found in Germany (de Souza Pereira and Kuch, 2005), whereas PCB concentrations almost 100 times higher have been found in sewage sludge in Brazil (de Souza Pereira and Kuch, 2005). Many researches confirm the presence of PCBs in sewage sludge and organic waste, which should be utilized (Blanchard et al., 2004; Brändli et al., 2007; Siebielska 2008). Two biological methods are generally used: composting and the anaerobic digestion. Rosinska and Dabrowska obtained decrease of PCB content over 50% during anaerobic digestion of sludge with smaller initial concentration of PCB, and over 60% in the sludge highly contaminated with PCB (Rosińska and Dąbrowska, 2014). Chang et al.

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during their studies on the anaerobic digestion of sewage sludge observed the dechlorination rate within the range of 0.063–0.025 mg L^{-1} day^{-1}. It depended on the PCB congener and the type of sludge (Chang et al., 1999). Therefore it can be assumed that the decrease of PCBs concentration during biological treatment strongly depends on the degree of chlorination of individual congeners. Brändli et al. (2007) conducting composting process of municipal waste in piles found that the concentration of higher chlorinated PCBs decreased while the concentration of lower chlorinated PCBs increased. Different relationship was observed by Michel et al. (2001) during research on the composting process in the remediation of soil contaminated with PCBs. The decrease of the lower chlorinated PCBs concentration due to biodegradation was over 40% and less than 1% due to volatilization.

Processes which may influence PCBs concentration during composting and anaerobic digestions are: sorption, evaporation, biodegradation, chemical treatment and other. It is difficult to determine which of those processes is the most influential. The PCBs biodegradation mechanism under both aerobic and anaerobic conditions is fully described in the literature (Tartakovsky et al., 2001; Borja et al., 2005; Field and Sierra-Alvarez, 2008; Furukawa and Fujihara, 2008).

The comparison of two biological methods of biowaste treatment is difficult because different processes of biological and chemical decomposition of organic matter take place under aerobic and anaerobic conditions. First of all, the duration of the two processes is different. In the case of anaerobic digestion it is 14–28 days while in the case of composting it is 90–180 days.

In the presented study changes of the PCBs concentrations were analyzed during anaerobic digestion and composting of municipal waste. But the main aim of our research was comparison of those methods based on kinetics of the PCBs degradation. Only constants of the process rate k [day^{-1} or kg mg^{-1} day^{-1}] of the same order were compared. Additionally the effectiveness of the PCBs degradation during composting and anaerobic digestion was compared.

Low nitrogen content in the municipal waste significantly limits application of biological methods. Addition of sewage sludge, rich in nitrogen, increases the effectiveness of methods of biological stabilization (Sidełko et al., 2011). This is the reason why the mixtures of sewage sludge and organic fraction of municipal waste were examined in the research. The composting and anaerobic digestion were carried independently in two separate reactors.

2. Experimental details

2.1. Bioreactor batches

Digested sewage sludge was collected from the sewage bioreactors in a municipal wastewater treatment plant that receives c.a. 40 000 m^3 of wastewater per day. The sludge had a moisture content, after dehydration, of 75%. The analyzed PCB's concentrations in the material were ⟨ 0.010 ± 0.001–1.000 ± 0,086 ⟩ mg/kg.

The municipal waste organic fraction came from a waste disposal facility that processes about 45 000 mg of municipal waste per year. The unseparated waste was first passed through a 4 cm sieve and then through a 2 cm sieve. The organic fraction, which had a moisture content of about 50%, was collected. The analyzed PCB's concentrations in the material were < 0.030 ± 0.002–3.000 ± 0.101 > mg/kg.

In the first two bioreactor cycles (cycles 30I and 30II), the materials used in the bioreactors contained 30% municipal organic waste and 70% sewage sludge, and the material used in the next two cycles (cycles 50I and 50II) contained 50% municipal organic waste and 50% sewage sludge. The material used in the last two cycles (cycles 70I and 70II) contained 70% municipal organic waste and 30% sewage sludge. The volume of each batch was 50 dm^3.

2.2. Composting

Six composting cycles were conducted in a static 60 dm^3 bioreactor with forced aeration (30I c, 30II c, 50I c, 50II c, 70I c, 70II c). An air supply flow rate of 2.5 dm^3 min^{-1} was provided during the first composting stage, then the temperature rose, reaching a maximum of 60 °C. The air supply flow rate was decreased to 0.5 dm^3 - min^{-1} during the compost maturing stage. Each composting cycle lasted 182 d. The material that was composted was a mixture of dehydrated sewage sludge and the organic fraction of municipal waste. The surface area to volume ratio of the composted material was 15.7: 1 m^2 m^{-3}.

The sample collection frequency depended on the intensity of the composting process at a particular time. Samples were collected every 7 d for the first four weeks, then every 14 d for the next six weeks, and every 21 d for the next 12 weeks, and, following that, the last sample was collected 28 d later. Total nitrogen, total organic carbon and the PCBs concentrations were determined in collected samples.

2.3. Anaerobic digestion

Anaerobic digestion was conducted in a bioreactor with a capacity of 50 dm^3 (relative diameter 40 cm). The bioreactor was equipped with a 4 paddle stirrer that followed a 15 min stirring (at 10 rpm) then 30 min non-stirring cycle throughout the digestion process. A constant temperature was maintained using a heater that was controlled by a thermostat. There was a tap at the bottom of the bioreactor to allow samples of the mixture to be removed during the digestion process. Biogases were collected for analysis through a valve in the lid of the bioreactor.

Mesophilic conditions were maintained during the anaerobic digestion process, and the temperature was maintained at 39 °C. The methanogenesis stage lasted around 21 d. The material that was digested was a mixture of dehydrated sewage sludge and the organic fraction of municipal waste. Digested sludge was used as the inoculum.

Six anaerobic digestion process cycles were performed, using different sewage sludge and municipal waste organic fraction mixing ratios (30I ad, 30II ad, 50I ad, 50II ad, 70I ad, 70II ad). All of the other parameters were the same for all six cycles.

Samples were collected from the fermenting mixture every 2 d during the first week and every 3 d for the last two weeks of each digestion cycle. Total alkalinity, the volatile fatty acids (VFA) and the PCBs concentrations were determined in collected samples. Biogas sampling frequency was the same as the frequency of collection of the mixture samples.

2.4. Physical and chemical analyses

2.4.1. Total nitrogen

The total nitrogen concentrations were determined in fresh samples using an elemental analysis technique. The nitrogen-containing compounds were decomposed by catalytic oxygen combustion in a helium atmosphere at 900 °C. Nitrogen and nitric oxide were produced in that process, and the nitric oxide was then reduced to form nitrogen at a temperature of 830 °C. The analysis was performed using a varioMAX macro CN analyzer system (Elementar, Hanau, Germany) with a thermal conductivity detector (AN-A-220205-E-01).

2.4.2. Total organic carbon

The total organic carbon concentrations were determined in dried samples using elemental analysis. The carbon compounds in a sample were decomposed by combustion with oxygen in a helium atmosphere at 900 °C. The analysis was performed using