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Transport of PCBs with Leachate Water from the Contaminated Soil

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Abstract Contaminated soil was taken from the area around the damaged capacitor of an electrical transformer station in Zadar (Croatia) and a phytoremediation experimental field was constructed with lysimeters. The levels of PCBs in the leachate water samples were measured nearly 3 years. The experiment was conducted under natural climatic conditions, through hot summers and mild winters. Although the amounts of leachated PCBs from the polluted soil were relatively small, their concentrations in leachate water samples from 1,500 to 29,000 ng L^{-1} substantially exceeded the maximum allowed concentration of the total PCBs in the fishponds of Croatia (1 ng L^{-1}).

Keywords PCBs · Research phytoremediation field · Leachate water · Croatia

Soil was contaminated by PCBs from a damaged electrical transformer station in Zadar (Croatia) during the autumn of 1991, when condenser oil was spilled over the ground. The level of PCBs in the soil around the damaged condenser indicated significant contamination (Picer et al. 2003, 2004). The soil was excavated from the polluted site and a phytoremediation experimental field was constructed with lysimeters (Picer et al. 2006; Hodak Kobasić et al. 2005). During the exposure of a field to atmospheric precipitation, the water goes toward the deeper soil layer, and thereby the pollutants are transported by the processes of desorption in the percolating water (Jones et al. 1989). The aim of the current study was to determine the leaching behavior of PCBs through thin layer of karstic soil over an extended period of time under natural climatic conditions.

Materials and Methods

Contaminated soil was collected from the area around the damaged capacitor. The homogenized soil was divided into ten plots (1 m × 3 m each) which were prepared about 10 m distance from the polluted area (Picer et al. 2004). At the beginning of the experiment, the soil in the plots was 15-cm thick. The lysimeters (70 cm × 70 cm × 20 cm) were made of stainless steel and placed in the soil plots at an angle in order to allow the collection of the water in the bottles. Each lysimeter ended with a screen and a 10 cm extension connecting it to a 10-L collection bottle. To exclude the effects of sunlight, the collection bottles were placed in wooden containers buried at a depth of 70 cm from the surface and 15 cm below the experimental plot (Fig. 1). Every lysimeter was filled with a 5 cm layer of gravel to promote water drainage and a 15 cm layer of homogenized contaminated soil. Each plot had three lysimeters. The experimental plots were seeded in September 2004 with different plant species, but during the experimental period various types of weeds grew and prevailed. At the beginning of each leaching experiment, a mixture of n-hexane and methanol (3:1, respectively) and 1 L of tap water was added to every collection bottle. At least 7 L of leachate water were collected in every collection bottle. The periods of sampling were variable, depending on the intensity and frequency of raining.

PCB stability in leachate water was tested in order to determine the quantity of PCBs (Aroclor 1248 equivalents) that remained in the glass bottles after 1 month under natural field conditions. Each analysis was performed in

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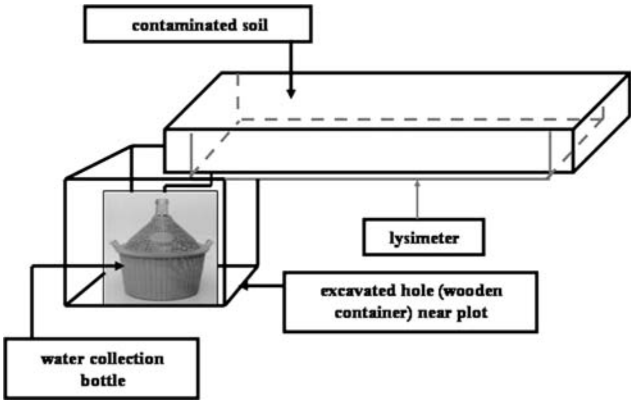


Fig. 1 The position of the lysimeter in relation to the collection bottle

triplicate. Glass funnels were filled with a layer of gravel and a layer of uncontaminated soil from the surrounding area, and placed over the glass bottles to replicate as closely as possible the field conditions with the lysimeters. Five liters of tap water were passed through the funnels and 5 µg of Aroclor 1248 in n-hexane/methanol solution (3:1) was added to glass bottles. The bottles with Aroclor 1248 and funnels were then placed in wooden containers and left for a month. The average values of the PCB content in each of the bottles were expressed as the sum of seven key PCB congeners (ΣPCB_7; IUPAC No.: PCB 28, PCB 52, PCB 101, PCB 118, PCB 138, PCB 153, and PCB 180). Recovery of ΣPCB_7 in a winter month was about 87% of the initial content while in a summer month the recovery was about 62%.

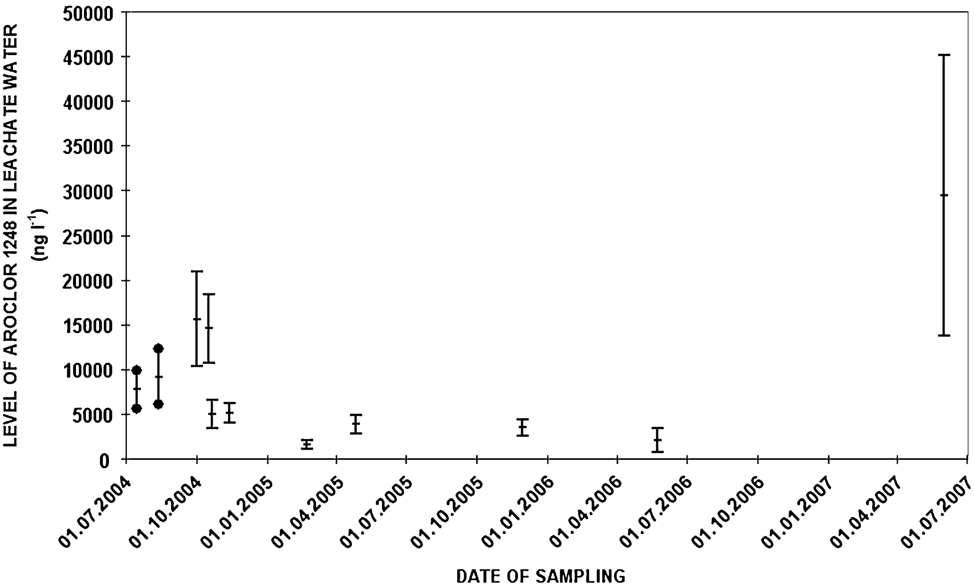
Water samples were filtered prior to analysis due to the substantial particle content. n-Hexane/acetone solvent mixture (1:1) was used for the extraction of the filter paper with filtered solid particles in an ultrasonic bath (two times for 30 min). Prior to the extraction of the leachate water filtrate, NaCl was added until saturation and acidified with HCl (36%) to obtain pH of 2. PCBs were double-extracted from the aqueous phase by liquid/liquid partitioning with the addition of 100 ml of n-hexane. The analytical method used to purify the obtained extracts included filtration through a 3-cm high column of Na_2SO_4 anhydrous and purification through an Al_2O_3 column. Na_2SO_4 anhydrous and Al_2O_3 were before application pretreated. Anhydrous Na_2SO_4 was heated for 12 h at 400°C and stored in a glass container in desiccators. The Al_2O_3 (Brockmann activity I) previously had been prepared by heating at 500°C for 12 h and partly deactivated by adding five percent distilled water by weight. Elution was performed with 15 ml hexane. The elutes were analyzed using the Agilent Technologies 6890 N network GC system, equipped with an electron capture detector (ECD) containing a cell plated with 63 Ni. The column used was a high resolution glass capillary HP-5 (cross-linked 5% Phenyl methyl siloxane), 30 m × 320 µm × 0.25 µm film thickness. The carrier gas was nitrogen. The experimental conditions of the instrument were as follows: injector temperature 523 K, detector 623 K, and the temperature program of the column oven 333–503 K.

The detection limit for the total PCBs in the leachates was 5 ng L^{-1}. Detailed descriptions of the methods used are published in numerous papers (Picer 2000; Hodak Kobasić et al. 2005).

Results and Discussion

As the leachate water was collected over periods much shorter than 1 month, our findings on the levels of PCBs in

Fig. 2 Aroclor 1248 (arithmetic mean with standard deviations) concentrations in the leachate water after irrigation (●-●) and rain period (I-I)



leachate water samples are fairly realistic. Fig. 2 shows total PCB concentrations in leachate water samples expressed as Aroclor 1248 equivalents of two irrigation and nine raining periods. Due to dry period at the beginning of the experiment (July and August 2004) several experimental plots were irrigated with tap water in order to study PCB leaching. The first irrigation was performed on three lysimeters from the same plot (15 July 2004). The second irrigation was done on two soil plots and six lysimeters (12 August 2004). The collection of leachate water samples was performed during the following rain periods: 13–30 September 2004, 1–16 October 2004, 17–20 October 2004, 21 October–11 November 2004, 27 January–21 February 2005, 8–26 April 2005, 26–28 November 2005, 5–23 May 2006, and 30–31 May 2007. The PCB concentrations for each rain period are presented as the arithmetic means with standard deviation measured in the leachate waters from all 12 lysimeters. The levels of PCBs during the initial period of the experiment were unevenly distributed among the leachate waters collected in various lysimeters. The reason for this could be the unsettled nature of the soil in the early phases of the experiment. The concentrations of the total PCBs in the leachate water collected after this initial stage were more uniform, except for February 2005, which showed somewhat lower values (1,555 ± 345 ng L^{-1}). As time proceeded, sorption and fixation of PCBs to the soil matrix increased and thus leaching losses were reduced. The total PCB levels in the leachate water collected in May 2007 were significantly higher. These findings could be explained by the appearance of shrinkage cracks in the karstic (clay loam) soil due to a long drought period. In May 2007 an intensive rainfall event occurred and high amounts of PCBs were rapidly transported with the water through the macropores, bypassing the thin layer of soil matrix in lysimeter.

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