



Distribution and release of 2,4,5-trichlorobiphenyl in ice

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Abstract

The distribution of persistent organic pollutants (POPs) in ice and the release of POPs from ice during ice melting have an important influence on the eco-environment and water quality of a river. Through laboratory simulation experiments, the distribution and release of 2,4,5-trichlorobiphenyl (PCB29) in ice and the partition coefficients of PCB29 in ice water at different temperatures, concentrations, and pH levels were studied. The results showed that, at different temperatures and concentrations, the concentration of PCB29 in ice increased progressively with depth. The modes of release of different concentrations of PCB29 from ice were obtained. A large amount of PCB29 was released rapidly in the first melting period, and then the remaining PCB29 was released uniformly. The pH value dominated both the distribution and late release of PCB29 in ice. In ice water, at different temperatures, concentrations, and pH levels, the majority of PCB29 entered the water, and a lesser amount remained in the ice. Finally, laboratory experiment results were verified with field investigations. A theoretical framework is provided by this research of the behavior of POPs in ice under different environmental conditions, but a more quantitative understanding of the behavior of POPs in ice will need to be developed through further laboratory studies combined with field investigations.

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Keywords: Persistent organic pollutants; 2,4,5-trichlorobiphenyl; Ice; Distribution; Release; Partition coefficient

1. Introduction

The 5464 km-long Yellow River originates in the northern part of the Bayankala Mountains at an altitude of 4830 m on the Qinghai-Tibet Plateau, in China (Huang et al., 1992; Fu, 1998), and is the fifth longest river in the world. In recent years, the river has been significantly polluted by contaminants. According to water quality monitoring results of the Yellow River in Inner Mongolia, water pollutants include persistent organic pollutants (POPs), described in the *Stockholm Convention on Persistent Organic Pollutants* (Yuan et al.,

2001), and the most typical POPs are organochlorine pesticides and polychlorinated biphenyls (PCBs) (Pei et al., 2010). POPs are a class of highly toxic substances with strong carcinogenicity, teratogenicity, and mutagenicity (Nicholas et al., 2001; Ricardo et al., 2005). There has already been a considerable amount of research on pollution of POPs in rivers (Fu et al., 2003; Rajendran et al., 2005; Fujii et al., 2007). As the Yellow River's freezing period in Inner Mongolia can last up to five months, some pollutants stranded in ice during the freezing period may cause secondary pollution in the river when ice starts to melt in the spring of the next year, in accordance with unique environmental chemical characteristics of rivers in the freezing period (Terry and Spyros, 2002). Therefore, study of the behavior of POPs in ice plays an important role in understanding the water quality problems of the Yellow River.

There have been some studies on organic contaminants in ice and snow. Melnikov et al. (2003) observed concentrations

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of POPs in ice and snow in the Ob–Yenisey River Watershed and concluded that the spatial analysis results of data on POPs for the watershed are in most cases similar to measurements carried out in the far north of Canada. Gustafsson et al. (2005) provided a coherent and reliable set of observations on the concentrations of PCBs in particulate and dissolved forms in snow and ice, showing that ice is not a significant medium for long-range transport of POPs in the Arctic. Pucko et al. (2010) presented evidence that both geophysical and thermodynamic conditions in sea ice are important in determining pathways of accumulation or rejection of hexachlorocyclohexanes. Further study by Meyer and Wania (2008) focused on organic contaminant amplification during snowmelt, concluding that there are essentially two patterns of organic contaminant enrichment in snow meltwater: one leads to preferential elution with the early meltwater fractions, and the other leads to enrichment of particle-sorbed substances in late meltwater fractions. These results provide an important starting point for any in-depth discussion of POP behavior in ice. However, these findings relate only to POP behavior in ice and snow under specific environmental conditions. Also, the actual ice environment is constantly changing, so it is necessary to investigate POP behavior in ice under different environmental conditions.

Based on laboratory simulation experiments, this study was meant to provide results on distribution and release of 2,4,5-trichlorobiphenyl (PCB29) in ice and partition coefficients of PCB29 in ice water at different temperatures, concentrations, and pH levels, and to verify these results with field investigations. It is hoped that this paper will provide a theoretical basis for studies on the impact of distribution and release of POPs in ice on water environments under different environmental conditions.

2. Materials and methods

2.1. Experimental methods

The PCB29 solution was prepared with distilled water and a PCB29 standard solution (0.005 mg/mL, with acetone as the solvent) after a long period of shaking. We then put the stainless steel barrel filled with PCB29 solution in the device (Fig. 1), which can create experimental conditions close to the freezing condition of natural rivers: freezing from top to bottom. Finally, the solution was frozen in a cryostat, which can control the temperature.

The experiment was organized as follows:

(1) Distribution experiment: One liter of PCB29 solution with a certain concentration and pH value (regulated with NaOH and HCl) was prepared, and completely frozen at a certain temperature. Then, the ice core was cut to three layers from top to bottom, melted, and analyzed.

(2) Release experiment: One liter of PCB29 solution with a certain concentration and pH value was prepared, and completely frozen at -25°C , then melted in the laboratory. Two hundred milliliter of meltwater was collected for every sample during melting and five water samples were analyzed.

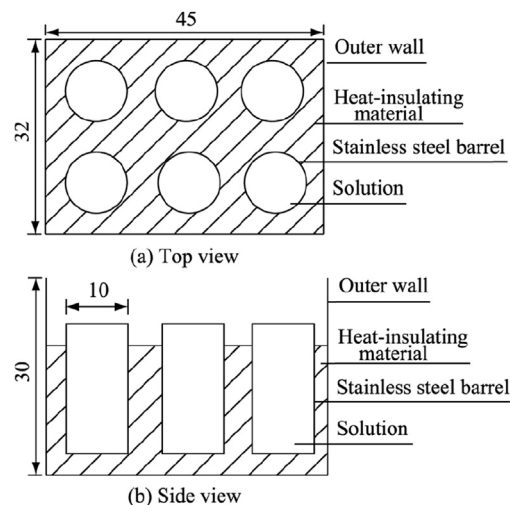


Fig. 1. Sketch of experimental device (units: cm).

(3) Determination of ice-water partition coefficient: One liter of PCB29 solution with a certain concentration and pH value was prepared, and frozen at a certain temperature. The ice-to-water volume ratio was kept at about 1:1. Then ice and water were separated for analysis.

According to these methods, the distribution and release of PCB29 in ice and partition coefficients of PCB29 in ice water at different temperatures, concentrations, and pH levels were investigated.

2.2. Field sample collection

From January 4, 2012 to February 21, 2012 (the freezing period), ice and water samples were collected in the Toudaoguai section of the Yellow River. During the sampling period, the average temperature was -5°C , the average pH value of ice was 7.6, and the average thickness of ice was 46 cm. Ice samples at 20 cm from the surface and 20 cm from the bottom were collected, and then placed in clean stainless steel barrels. Two-liter water samples under the ice were collected and put in brown glass bottles.

The river ice began to melt between February 22 and February 24, 2012 (the average temperature was 3°C). Three two-liter water samples were collected from the shore on February 24, March 13, and March 31.

All samples were transported to the laboratory for analysis.

2.3. Concentration analysis

2.3.1. Extraction of PCB29

This experiment used the solid phase extraction (SPE) method to extract PCB29 from the solution. The specific steps were activating the SPE column, solution-adding, drying, and eluting.

Activating the SPE column consisted of leaching the C18 column at a rate of 1 mL/min with 5 mL acetone, 5 mL hexane, and 5 mL distilled water in turn, to keep the column moist when adding solution. Solution-adding occurred when

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