



Sorption behaviors of a persistent toxaphene congener on marine sediments under different physicochemical conditions



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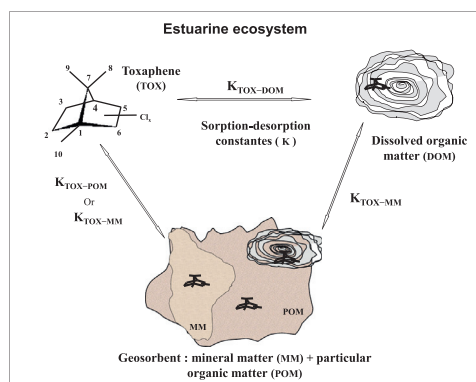
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HIGHLIGHTS

- We reported the sorption and desorption coefficients of a toxaphene congener.
- The B7-1450 was studied under different temperature and salinity conditions.
- We model sorption data to understand the B7-1450 distribution in estuarine system.
- The B7-1450 mobility is higher in marine system than in freshwater medium.
- Climate change could enhance the bioavailability of toxaphene congeners.

GRAPHICAL ABSTRACT



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ABSTRACT

Sorptive processes are important parameters affecting the mobility, availability and fate of persistent organic pollutants (POPs), such as toxaphene, in aquatic systems. The sorption and desorption behaviors of the B7-1450, a stable toxaphene congener in environment, on marine sediment was studied under different temperature and salinity conditions to better understand the B-1450 distribution in estuarine systems. The data were fitted to different sorption models to characterize sorption behaviors by evaluating sorption coefficients and sequestered fraction of B7-1450 on sediments. High carbon-normalized sorption coefficients (K_{oc}) of the B7-1450 were observed with values ranging from 3.2×10^4 to $6.0 \times 10^4 \text{ mL g}^{-1}$ under experimental conditions. The data showed an increase of B7-1450 sorption coefficients with the salinity and a decrease with temperature. These investigations indicate that B7-1450 is three times more sequestered on sediments in cold (2°C , 30 psu) than in warm marine conditions (20°C , 30 psu). These results suggest that the mobility and bioavailable of B7-1450 or other POPs from the sediments could be less important in cold marine comparatively in warm marine and warm freshwater media. As a result of climate changes, the warming of mid and high latitudes coastal waters could enhance the mobility of POPs.

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1. Introduction

The toxaphene has been one of the most widely used chlorinated pesticides and is among the 12 original priority persistent organic pollutants (POPs) (Saleh, 1991). The technical toxaphene is a complex mixture of several hundred chlorobornane and others chloroterpene congeners (Vetter and Oehme, 2000). Despite the restriction of toxaphene use three decades ago, residues of chlorobornanes were still reported in water column, sediments and biological matrices of several ecosystems (Gouteux et al., 2003, 2005; Muir et al., 2006). Low levels of heptachlorobornanes, such as B7-1450, were found in biological matrices (15% of all detected toxaphene) whereas heptachlorobornanes are predominantly present in sediments (25–55%) (Braekevelt et al., 2001; Muir et al., 2006). The B7-1450 congener, a heptachlorobornane, could account for 15–20% of all congeners detected in sediments (Pearson et al., 1997; Gewurtz et al., 2007).

Interactions of hydrophobic organic compounds (HOCs), including toxaphene, with sediments represent major processes that control their distribution and bioavailability in aquatic systems (Gagné et al., 2011). To our knowledge, sorption interactions of toxaphene with particulate matter, such as sediments, have received limited attention although 1.3×10^6 tons of this pesticide has been spreaded in the environment between 1950 and 1993 (de Geus et al., 1999). One study examining the sorption of a specific congener of toxaphene on sediments was published (Soubaneh et al., 2008). Therefore, toxaphene sorption studies onto sediments from St. Lawrence estuary will allow to better understand the availability and mobility of these compounds in the estuarine ecosystem.

Sorption behaviors of HOCs on sediments are commonly examined through sorption kinetic studies (Huang et al., 1996). Sorption kinetic processes of HOCs on sediments or soils occur on wide time scales according to the feature of the organic matter (OM) or the mineral content. The OM in sediments or soils could have amorphous and condensed states with low- and high-energy sorption sites, respectively (Gunasekara and Xing, 2003). The first sorption step is generally fast, occurring on the order of minutes to days, and reflects sorption interactions associated with amorphous OM or with mineral surfaces (Weber and Huang, 1996). The initial sorption process is followed by a slow step, occurring on the order of weeks to years, and suggests HOCs sorption on condensed OM or diffusion into geosorbent small pores (Gunasekara and Xing, 2003).

Sorption–desorption isotherm studies allow understanding the sorptive behaviors of HOCs on sediments in equilibrium conditions (Limousin et al., 2007). The sorption process permits to evaluate the sorption coefficient (K_d^S) of HOCs on sediments in order to assess the affinity of HOCs for sediments. The desorption process determines the desorption coefficient (K_d^D) and allows estimating the HOCs fraction entrapped or sequestered in sediments. These “irreversible sorption” phenomena, named hysteresis, have been explained as the result of the HOCs molecules sequestration in OM or mineral surfaces meso-/microporous structures (Huang et al., 1998).

The sorption isotherms could provide further insight into sorption mechanisms involving sediments OM which is a dominant sediment constituent for sorption and sequestration of HOCs. A linear sorption was reported for HOCs mainly partitioned on low-energy sites available on sediments (Ran et al., 2003). In contrast, a nonlinear sorption was observed when HOCs were predominantly sorbed on high-energy sorption sites.

Salinity and temperature parameters can affect sorption–desorption behaviors of HOCs on sediments particularly in estuarine systems susceptible of being affected by climate change. Estuarine systems are also mixing zones for fresh riverine and coastal ocean water masses (Turner and Rawling, 2001). Water

from river can be a potential source of toxaphene to estuarine systems. Raff and Hites (2004) estimated up to 1000 kg the amount of toxaphene released in 2002 into the Gulf of Mexico from the Mississippi river. The salinity and temperature changes are expected to be important in the sequestration and bioavailability of toxaphene in estuarine ecosystems. An increase of the salinity of the water is expected to decrease the HOCs solubility and enhance OM hydrophobicity of the sediment with an enhancement of HOCs sorption on sediments (Tremblay et al., 2005). In contrast, an increase of temperature is expected to enhance the solubility of HOCs in the water and to decline the sorption capacity of the sediment due to enhancement of the low-sorption sites available on the sediment OM (Zhao et al., 2003).

The aim of this work was to study the sorption behavior of the B7-1450 toxaphene congener as a model molecule of the sorption of toxaphene or other POPs on a marine sediment. Particular emphasis was placed on: (i) the sorption kinetics examination of the B7-1450; (ii) the characterization of the sorption and desorption isotherms using sorption models; and (iii) the investigation of salinity and temperature effects on the sorption of B7-1450 within the range of concentrations found in estuarine systems.

2. Materials and methods

2.1. Geosorbent

Surface sediments were collected with a box corer in the Gulf of St. Lawrence (GSL) (Canada) (49°24.0'N; 64°44.8'W, depth: 386 m). The first 5 cm of the core were sampled and freeze dried. The particle size of the GSL sediment was characterized (Table 1) using a Beckman Coulter LS 13320 and the elemental composition was determined using CHNO analyser (Costech 4010). Fulvic acids, humic acids and humin substances were obtained by extracting the sediment using the method described by Swift (1996). The dissolved organic carbon (DOC) released from sediments during the sorption experiments was analysed with a Shimadzu TOC-5050 analyser (Table S3). Analyses of organochlorine compounds contents in the sediment showed levels below detection limits for B7-1450 ($<3.4 \text{ ng g}^{-1}$ dry weight), PCB-55 and PCB-171 congeners ($<0.2 \text{ ng g}^{-1}$ dry weight).

2.2. Sorbate and chemicals

The toxaphene congener, 2-*exo*,3-*endo*,5-*exo*,8,9,10,10-heptachlorobornane, B7-1450, was prepared and purified according to the procedure described by Nikiforov et al. (1995). The B7-1450 had purity greater than 98% as determined by gas chromatography

Table 1
Selected properties of the Gulf of St-Lawrence sediment.

Sand (%) (62.5 μm –2.0 mm)	6.5
Silt (%) (4.0–62.5 μm)	81.3
Clay (%) (<4.0 μm)	12.2
f_{oc} (%) ^a	2.0
N (%)	0.3
H (%)	0.9
O (%)	3.9
C/N	8.8
O/C	0.9
H/C	1.6
(O + N)/C ^b	1.1
Fulvic acids ^c	10.3
Humic acids ^c	4.0
Humin ^c	25.9

^a Organic carbon.

^b Polarity index.

^c Humic substances (mg g^{-1} of sediment).

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