

Evolution rates and PCB content of surface films that develop on impervious urban surfaces

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Received 24 October 2007; received in revised form 18 January 2008; accepted 19 January 2008

Abstract

Surface films on impervious surfaces mediate the fate of organic contaminants in urban areas. Using sheltered 3-mm diameter glass beads as a surrogate impervious urban surface, studies in downtown Toronto showed that surface films developed at a consistent rate of $\sim 1.6\text{--}2.6\text{ nm day}^{-1}$. Linear film growth was observed up to and including the last day of three studies of 89-, 273-, and 84-day duration. Total PCB content (sum of 15 PCB congeners) also increased linearly, at a similar rate as the surface film. A complementary study using unsheltered beads showed that up to 80% of film mass and PCB content were removed by film wash-off during rain events. The film growth rate on beads exposed at a rural site, 200 km northeast of Toronto, was $\sim 12\text{--}20$ times lower with PCB burdens that were $\sim 7\text{--}22$ times lower compared to Toronto. Based on these findings, it is estimated that approximately 56–226 g of PCBs are captured by a 70 nm thick film covering just the horizontal surface area of Toronto. This is a substantial quantity of PCB that is available for rapid surface–air exchange and wash-off.

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Keywords: Surface films; PCBs; Impervious surfaces; Urban environments

1. Introduction

Surface films have been found to coat impervious surfaces, thereby changing their functionality with respect to chemical dynamics. The organic content of the film, which is hypothesized to accumulate via condensation of gas-phase species and the deposition of particulate matter with its constituent organic species, allows the film to act as a temporary

sink for semi-volatile compounds (SOCs). These compounds can undergo temperature-dependent partitioning, allowing volatilization back into the atmosphere when conditions are favorable (Gingrich et al., 2001; Diamond et al., 2000, 2001). Because of the large surface area of the film, which can exceed that of particulate matter in urban settings, surface–air exchange dynamics are increased. This may help to buffer air concentrations of SOC and increase their mobility (Priemer and Diamond, 2002). The films will also be washed off during precipitation events (Labencki et al., submitted for publication) and contribute to contaminant loads to urban surface waters.

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This phenomenon is similar to that described by Mahler et al. (2005) who investigated the contribution of sealed coal-tar pavements as a source of PAHs to the urban watershed. Unlike soils, which are, in effect, long-term sinks for chemicals, surface films serve as a dynamic sink/source of chemicals.

Sources of particulate matter and surface films are highly influenced by anthropogenic activities, including vehicle emissions, stationary and mobile fuel combustion, cooking and cigarette smoking (Rogge et al., 1993a, 1994; Lam et al., 2005). Biogenic sources of organic compounds include plant waxes, other vegetative debris and aero plankton while inorganic sources include crustal material (e.g. mould spores, bacteria) (Rogge et al., 1993b; Liu et al., 2003; Lam et al., 2005). The organic fraction of the film may only comprise 4–5% of the mass, with the greatest mass contribution presumed to be silicates followed by other inorganic species such as sulphate and nitrate (Lam et al., 2005). Due to the abundance of emissions and geographic gradients in particulate matter concentrations, film thickness varies greatly from urban to rural locations and could differ based on seasonal or diurnal variations (Gingrich et al., 2001; Liu et al., 2003).

To evaluate the influence of surface films on impervious surfaces to chemical dynamics and mass balances for SOCs, it is important to understand how the film evolves and how SOCs accumulate. This paper presents the results from experiments documenting surface film development using 3-mm glass beads as a surrogate impervious surface. We investigated the accumulation of polychlorinated biphenyls (PCBs) as a class of SOCs, the effect of meteorological factors on film growth, and compared film growth rates in an urban vs. rural site. PCBs are ubiquitous in the urban environment, originating from open sources such as building sealants and exterior paints, and from closed sources such as in-use dielectric fluid in capacitors and transformers and stored waste (e.g., Herrick et al., 2004, 2007).

2. Materials and methods

2.1. Sampling

To characterize film growth, surface films were allowed to accumulate on 3-mm diameter Pyrex glass beads (Fisher Scientific) during five sampling campaigns. Glass beads were chosen as a surrogate

impervious surface to maximize sampling surface area. Field study locations included the roof of a three-story building (PGB) in downtown Toronto and a rural sampling site, Dorset. Table 1 summarizes the sampling campaigns conducted over 2 years in this study.

In downtown Toronto, there were two non-consecutive sampling periods of 89 days from September to December 2003 (PGB-1) and 273 days from July 2004 to April 2005 (PGB-2). Thirty samples of glass beads, arranged as a monolayer, were set out on top of the PGB building and exposed horizontally to ambient air from all directions, but sheltered from direct precipitation, deposition, and radiation (Fig. 1). Each sample is composed of one aluminum tray of either 75 g of beads in PGB-1 study or 80 g of beads in PGB-2 study (corresponding to approximately 2250–2400 individual 3-mm diameter glass beads, or an equivalent total surface area of 636–679 cm²). During the initial 89-day study, a sample was collected every 1–2 days; during the second sampling period, a sample was collected every 1–2 weeks. Samples were collected randomly, where each sample tray was numbered and the collection order based on the results from a random number generator.

Differences in film evolution rates and SOC content of urban vs. rural samples were investigated by setting up parallel bead samples at Dorset, Canada (Fig. 1b), and at PGB, again over two separate time periods: winter 2004/2005 (November 2004–January 2005) and spring/summer 2005 (March–June 2005). Dorset is located approximately 200 km northeast of Toronto, at 45°14N, –78°54W on the southern margin of the Precambrian shield near the southwestern tip of Algonquin Provincial Park. The sampling site was in a compound set up and administered by the Ontario Ministry of the Environment, located outside of the town limits. The site was away from the compound's parking lot, with little direct exposure to local anthropogenic sources of particles. This urban/rural study is separately referred to as PGB-3 (urban samples) or Dorset (rural samples) (Table 1).

Finally, to investigate the impact of meteorological factors on film evolution rates and the consequence of precipitation events on film dynamics and composition, a further sampling study was set up at PGB (referred to as PGB-4). In this study, 24 trays of 3-mm diameter glass beads were set out on

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