

Partitioning of mono- and polycyclic aromatic hydrocarbons in a river sediment adjacent to a former manufactured gas plant site

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Abstract

The equilibrium distributions, between water and coal-tar contaminated sediment, of 16 monocyclic and polycyclic aromatic hydrocarbons were measured and evaluated for consistency with a Raoult's Law-based quantitative relationship. The quantitative relationship calculates the pore water concentration as the product of the aqueous solubility (or for compounds that are solid at room temperature, the aqueous super-cooled liquid solubility) and the mole fraction concentration of the compound within the liquid coal tar. Sediment was collected at five locations at two depths within a 120 m stretch of a river adjacent to a former manufactured gas plant, and all samples contained non-aqueous phase liquid (NAPL) coal tar. Although the amount of coal tar varied between samples by over an order of magnitude, the Raoult's Law-based NAPL–water partition coefficients for each monocyclic or 2- or 3-ring polycyclic aromatic hydrocarbon measured in this study generally varied within a factor of 2 over all sediments.

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1. Introduction

Manufactured gas plants (MGPs) in North America and Europe were one of the principal suppliers of energy in the 1800s and early 1900s. These plants manufactured gas from coal and oil and generated a variety of process wastes such as tars, spent oxides, ash, sludge, ammonia liquors and lampblack. The 1940s and 1950s saw the clo-

sure of these plants due to the availability of natural gas; however, wastes generated at MGPs often remain in the soil and groundwater at these sites. The major waste product was coal tar, largely consisting of a mixture of mono- and polycyclic aromatic hydrocarbons (MAHs and PAHs, respectively). In the US alone, there are 1000–2000 contaminated MGP sites, making coal-tar contamination a significant environmental problem (see Luthy et al., 1994, for a general review).

Most studies on risk assessment (Brown et al., 1999; Stroo et al., 2000), mass transfer (Peters and Luthy, 1993; Mukherji et al., 1997; Mahjoub et al., 2000), partitioning (Lee et al., 1992; Lane and Loehr, 1995), and remediation (Luthy et al., 1994) of coal-tar constituents

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at MGP sites have focused on coal-tar contaminated soil; however, many former MGP sites are located adjacent to creeks or rivers, resulting in PAH contamination of the sediment within these streams. When partitioning is described in the literature involving PAHs in sediments; however, the scenario invariably involves PAHs sorbed to sediment particles at concentrations below their saturation limit. At the MGP site that is the focus of this study, this is not the case, as the sediment in the adjacent river contains a non-aqueous phase liquid (NAPL) spread along several hundred meters of river reach to a depth of at least two meters in soft sediment deposits. In this case, phase distribution, and hence pore water concentration, is controlled by the NAPL composition.

Several previous studies have reported on the partitioning of PAHs between liquid coal tar and water, and have reported on the accuracy of Raoult's Law as a model to estimate aqueous phase concentrations from coal-tar composition (Lane and Loehr, 1992, 1995; Lee et al., 1992; Peters and Luthy, 1993; Woolgar and Jones, 1999; Mahjoub et al., 2000). Raoult's Law assumes ideal behavior of each component within the NAPL, and with the additional assumption that the activity of each aqueous phase component is not affected by other aqueous phase components (i.e., infinity dilution), the following equation can be derived:

$$C_w = C_o \cdot V_o \cdot S, \quad (1)$$

where C_o and C_w (mol/l) are the NAPL and aqueous phase PAH (solute) concentrations, respectively, V_o (l/mol) is the molar volume of the NAPL, and S (mol/l) is the solute's pure aqueous phase solubility. For solutes that in their pure phase are solids at room temperature, S is the hypothetical super-cooled liquid solubility. Among the previous studies listed above, differences of opinion exist among the authors on the accuracy of this equation and the appropriateness of its underlying assumptions in predicting equilibrium aqueous phase concentrations from coal-tar composition and pure phase aqueous solubility data. In applying this equation to coal-tar contaminated sediment, an addition problem arises: Due to the silty, organic nature of the sediment, it is impossible to completely remove the NAPL from the sediment to quantify its volume and mass—necessary parameters for calculating mole fraction concentrations. In these previous studies, the liquid coal tar has been removed from the soil or recovered from tanks at sites. In this study, an alternative approach in applying this equation is used. To this end, we report on the extent and composition of coal-tar contamination within the sediment along a river reach adjacent to a former MGP site, and test Eq. (1) and its assumptions as sufficient theory in determining the equilibrium distribution of MAH and PAH components between the NAPL contaminated sediment and water.

2. Materials and methods

2.1. Materials

Sediment cores (4-in. diameter \times 2.4-m) were collected with a P3 Vibrocorer (Rossfelder Corp.) at five locations within the small river adjacent to a former MPG site. Samples were collected within a 120 m reach of the river, with the closest two sampling locations approximately 22 m apart. Cores were sectioned in half, and each section (top 1.2-m and bottom 1.2-m) was mixed to form a composite sample. The composite samples were stored at 4 °C in metal containers. To ensure that identical samples were obtained from such large composites, each sample was homogenized prior to sub-sampling. Composite samples were labeled 1–5, T (top 1.2-m) and B (bottom 1.2-m), sequentially from upstream to downstream.

2.2. Experimental methods

Water and volatile organic content and ignitable contents were measured by heating the samples at 105 °C for 24 h, and at 500 °C for 3 h, respectively. The total organic carbon (TOC) content of each sample was measured with a Carlo-Erba 1108 elemental analyzer. Because of the sensitivity of the elemental analyzer and the large mass of carbon in each sample, 200–400 mg samples were pulverized with a Spex CertiPrep 6750 Freezer/Mill pulverizer, prior to weighing 4–5 mg samples into silver sample boats. This guaranteed that the small sample size was representative of the total. The water + volatile, ash, and TOC are reported in Table 1. Total organic matter (OM) was estimated by multiplying the TOC content by 1.7 as suggested by Chiou (2002). Non-volatile organic matter was assumed to be the difference between the masses remaining at 105 and 500 °C, and the volatile organic matter content was estimated as the difference between the total OM and non-volatile OM. These values are reported in Table 1 for all the sediments. Note that the volatile OM values (3.3–13.8% wet weight) suggest the presence of NAPL.

The 16 MAHs and PAHs that were the target analytes for this study are listed in Table 2 along with some important properties (Chiou et al., 1982; Lee et al., 1992; Schwarzenbach et al., 1993, 2003). A 16-PAH standard mixture was purchased from Ultra Scientific, and ethylbenzene, 1,3,5-trimethylbenzene, 1,2,4-trimethylbenzene, *m*-xylene, *p*-xylene and *o*-xylene were obtained from Aldrich Chemical Company. Methanol, dichloromethane and hexane were obtained from Fisher Scientific International, Inc.

To determine the concentrations of all 16 target analytes in each sediment sample, moist sediment (2 g) was extracted with 4 ml methanol and 11 ml dichloromethane on a rotary mixer at 60 rpm for 24 h. Each sample

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