



Translocation of fluoranthene in porous media by advancing and receding air–water interfaces



Maninder K. Chahal^{a,b}, James B. Harsh^b, Markus Flury^{a,b,*}

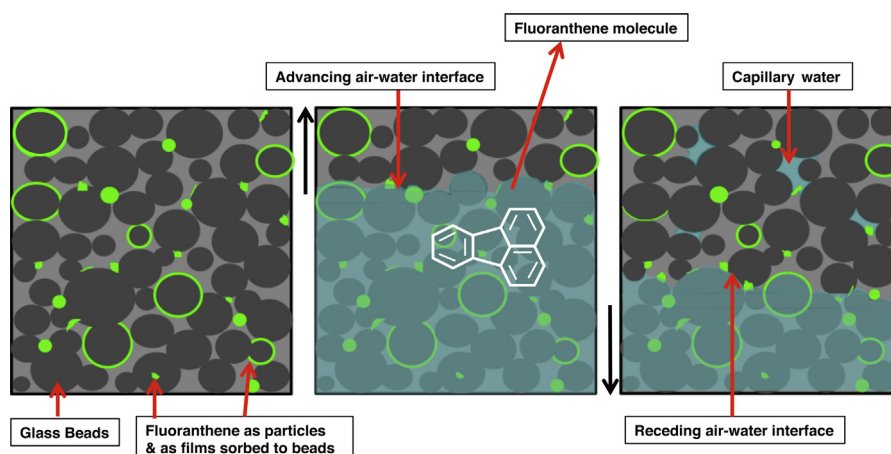
^a Department of Crop & Soil Sciences, Washington State University, Pullman, WA 99164, USA

^b Department of Crop & Soil Sciences, Washington State University, Puyallup, WA 98371, USA

HIGHLIGHTS

- Fluoranthene accumulates at air–water interface.
- Moving air–water interfaces translocate fluoranthene in porous media.
- Advancing air–water interfaces remove fluoranthene better than receding interfaces.

GRAPHICAL ABSTRACT



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ABSTRACT

Polycyclic aromatic hydrocarbons (PAH) generally have low water solubility and high octanol–water partition coefficients. In soils and sediments, PAH tend to partition to organic matter and mineral phases, and are, therefore, relatively immobile. However, moving air–water interfaces may scour PAH from solid phases and translocate PAH. The objective of our study was to assess the effect of moving air–water interfaces on the transport of fluoranthene, a four-ring PAH, in unsaturated porous media. A glass channel (i.d. 3.7 mm × 7.5 cm) packed with fluoranthene-contaminated glass beads (dia. 0.5 mm) was used as porous medium. Fluoranthene was initially present as species sorbed to the glass beads and as particulates precipitated out from solution. Capillary fringe fluctuations were simulated by repeatedly imbibing and draining the medium with water. Fluoranthene translocation was visualized by confocal microscopy and quantified by image analysis. Results showed that the first two cycles of capillary fringe fluctuations were the most effective in translocating fluoranthene; 25–50% of fluoranthene surface coverage was translocated from the glass channel. During imbibition, fluoranthene particles were detached from the solid surface, and attached to the advancing air–water interface. During drainage, fluoranthene was removed from the solid–liquid interface with the receding air–water interface. Repeated imbibition and drainage cycles, causing air–water interfaces to move through the porous medium, can be useful method to remove PAH from contaminated soils and sediments.

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* Corresponding author at: Department of Crop & Soil Sciences, Washington State University, Puyallup WA 98371, USA.
E-mail address: flury@wsu.edu (M. Flury).

1. Introduction

Polycyclic aromatic hydrocarbons (PAH) are a group of organic molecules containing two or more fused aromatic rings. Polycyclic aromatic hydrocarbons have low water solubility ($0.00094\text{--}242\ \mu\text{mol L}^{-1}$ at $25\ ^\circ\text{C}$) and high *n*-octanol water partition coefficients ($\log K_{ow}$ 3.37–6.04 at $25\ ^\circ\text{C}$) [1]. Many PAH are genotoxic, carcinogenic, and recalcitrant [2,3].

Polycyclic aromatic hydrocarbons are introduced into the environment by both natural processes and anthropogenic activities. Natural processes producing PAH include wild fires, volcanic eruptions, diagenesis of organic matter, and biosynthesis by bacteria, fungi and plants [4,5]. Anthropogenic activities include incomplete burning of fossil fuel, wood, solid wastes, and vehicular emissions [6]. When PAH are released to the environment, they can contaminate water and land resources. Due to their low water solubility, PAH tend to sorb and accumulate in soils and sediments [7] and in living organisms [8]. Nonetheless, there is the potential for PAH to leach through soils and to contaminate groundwater resources [9,10].

Various mechanisms have been proposed to identify the fate and transport of PAH in soils and sediments. In soils, PAH partition into organic matter [11] and sorb to clay minerals [12,13]. Polycyclic aromatic hydrocarbons are typically not very mobile in soils and sediments and do not readily leach through soil [14–16]; however, the presence of colloids or dissolved organic matter (DOM) can enhance the transport of PAH [17–19]. Facilitated transport of PAH in presence of DOM involves (1) sorption of PAH onto the soil matrix, particularly soil organic matter, (2) formation of complexes between DOM and PAH, (3) co-transport of the DOM-PAH complexes, and (4) sorption of the DOM-PAH complexes onto the soil matrix [19,20]. Biocolloids, such as mobile bacteria, can also facilitate the transport of PAH [21].

Transport of PAH in porous media has been studied mostly under saturated flow conditions [14,17,18,22,23] and only limited information is available on transport of PAH under unsaturated flow conditions [19,20,24,25]. Due to their hydrophobicity, PAH tend to accumulate at the air–water interface, e.g., at water droplets (dia. 14–200 μm) [26], and concentrations of PAH at the air–water interface can exceed bulk liquid phase concentrations by 2–3 orders of magnitude [27]. Elevated PAH concentrations have been reported from the air–water interface of seawater [28] and waste water lagoons [29].

The sorption of PAH at the air–water interface has implications for transport in porous media. Air–water interfaces can facilitate the transport of colloids, which are being attached to the interface by capillary forces [30–33], and we expect that this facilitated transport also occurs with PAH sorbed to the air–water interface.

Confocal laser scanning microscopy is a powerful technique to visualize spatial distributions of fluorescent substances, such as most PAH. Confocal microscopy has been used to investigate PAH spatial distribution in plant cells [34] and soils [22,35]. Confocal microscopy can likely also be used to study the interactions of PAH with air–water interfaces in unsaturated porous media, although we are not aware that this has been attempted yet.

The interactions and transport of PAH in unsaturated porous media are important processes for soils contaminated by oil or petroleum spills. Polycyclic aromatic hydrocarbons are also present in stormwater runoff, particularly from heavy-traffic roadsides, and such runoff often infiltrates into nearby soils or sediments. The objective of our study was to determine the effect of moving air–water interfaces on transport of PAH in porous media. Moving air–water interfaces occur commonly in soils and sediments during water infiltration and drainage. We selected fluoranthene as a representative PAH for our experiments because its strong fluorescence makes it a good candidate for the use of confocal

microscopy. We hypothesized that moving air–water interfaces facilitate the transport of fluoranthene through a porous medium.

2. Materials and methods

2.1. Overall experimental setup

Our experimental approach was to investigate whether moving air–water interfaces, typical for the capillary fringe region, can translocate fluoranthene contamination within a porous medium. We used a model porous medium made of glass beads contaminated with fluoranthene, and simulated capillary fringe fluctuations by repeatedly imbibing and draining the porous medium with deionized water. This caused advancing and receding air–water interfaces to move over the fluoranthene-contaminated glass beads. A water-saturated porous medium was used as a control to test the effectiveness of the moving air–water interfaces in removing fluoranthene. Fluoranthene removal was quantified *in situ* by confocal microscopy.

2.2. Chemicals and glass beads

Fluoranthene was obtained in powder form (98%, Lot No. MKBJ6602V, Sigma–Aldrich, St. Louis, MO). Fluoranthene is a four-ring PAH with water solubility of $1.28\ \mu\text{mol L}^{-1}$ at $25\ ^\circ\text{C}$ [36], octanol–water partition coefficient $\log(K_{ow})$ of 5.22 at $25\ ^\circ\text{C}$ [7], and excitation and emission wavelengths of 289 and 444 nm, respectively [37]. We dissolved fluoranthene in ethanol (97%) by adding excess amounts of the PAH to produce a saturated solution. The solution was equilibrated on a magnetic stirrer for 3 to 4 days and then centrifuged at 6237 RCF for 45 min [38]. The supernatant solution was then stored in an Erlenmeyer flask and covered with aluminum foil. The resulting solution should have a saturated fluoranthene concentration in ethanol of $92\ \text{mmol L}^{-1}$ at $25\ ^\circ\text{C}$ [39].

Glass beads with diameter of 0.5 mm (Part No. 11079105, BioSpec Products, Bartlesville, OK) were washed in ethanol and in a diluted HCl solution to remove organic and inorganic impurities, rinsed in deionized water, and air-dried [40]. Acid-washed glass beads were then soaked in fluoranthene-saturated ethanol solution in a glass Petridish for 3–4 days. As the ethanol evaporated, fluoranthene sorbed and precipitated onto the glass beads (Fig. S1, Supporting Information). Ethanol was used because the solubility of fluoranthene is much higher in ethanol than in water.

Advancing and receding contact angles of the air–water interface on clean glass beads, on fluoranthene-covered glass beads, and on fluoranthene particles were measured by using a manual goniometer (Model 147 50-00-115, Rame–Hart Instrument Co., Netcong, NJ). For the glass beads, a soda-lime microscopy glass slide (25 mm \times 75 mm, Fisher brand, Fisher Scientific) was used as a surrogate. Glass slides were rinsed with acetone, ethanol, and finally with deionized water. One glass slide was immersed into a fluoranthene-saturated ethanol solution in a Petridish, and the ethanol was evaporated in a laminar flow chamber. This produced crystallized fluoranthene particles and a fluoranthene-covered glass slide. The contact angles were measured by placing a drop of deionized water onto the slides with a glass syringe, such that the water drop was still attached to the syringe. Then, the goniometer platform was moved left and right to force the drop to slide over the glass slide, while the contact angle was then measured as the air–water interface advanced or receded. For the fluoranthene particle measurements, individual millimeter-sized particles were placed on a glass slide that was covered with double-sided tape. Advancing and receding contact angles were then measured

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