



How good are the predictions of mobility of aged polychlorinated biphenyls (PCBs) in soil? Insights from a soil column experiment

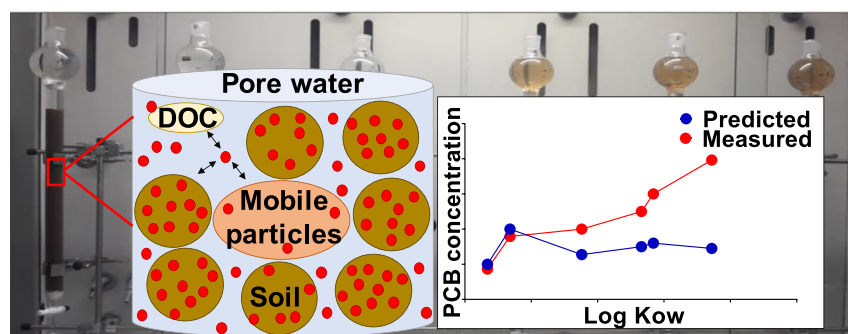
Chiara Maria Vitale, Elisa Terzaghi, Dario Zati, Antonio Di Guardo *

Department of Science and High Technology (DISAT), University of Insubria, Via Valleggio 11, Como, Italy

HIGHLIGHTS

- A soil column experiment was performed simulating variable environmental conditions.
- Contact time and temperature affected PCB fluxes associated to DOC/fine particles.
- DOC mediated transport of PCBs was not always linearly dependent on $\text{Log } K_{\text{ow}}$.
- Variation of saturation conditions determined peaks of contamination.
- PCB vertical movement with particles is a key feature to be included in fate models.

GRAPHICAL ABSTRACT



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ABSTRACT

A column leaching experiment was performed to evaluate the influence of some relevant environmental factors (soil/water contact time, temperature, saturation) on mobility of aged polychlorinated biphenyls (PCBs) in soil together with transport mediated by dissolved organic carbon (DOC) and mobile organic carbon (OC) coated fine particles/colloids. Consecutive fractions of leachates were collected after a variable pre-equilibration time (2, 5, 7, 48 days), using leaching solutions with different DOC content (tap water vs. Aldrich humic acid), in saturated vs. field capacity conditions and at different temperatures (25 °C vs. 15 °C). The data obtained were compared to the predicted values using a multimedia model (SoilPlusVeg) to evaluate model behaviour. Contact time and temperature determined a relevant effect on DOC and particle/colloid availability, with significant variations in leachate concentrations (up to 1 order of magnitude), typically overlooked by most environmental fate models. Results obtained at different temperatures show a modulation of the DOC/particles production with temperature and therefore the role of temperature changes in the environmental scenarios (e.g. seasonal variations). Transport of PCBs enhanced by Aldrich DOC was not linearly correlated to chemical hydrophobicity but revealed a threshold to $-\text{Log } K_{\text{ow}} 6.5$, likely because of the slow sorption kinetics of more hydrophobic chemicals. Additionally, variation of the saturation conditions (e.g. drying-wetting cycles) can determine contamination peaks at the beginning of an irrigation/rainfall event because of the soil/water equilibration. Model simulations, even when including DOC in the water phase, but not accounting for the particle/colloidal transport and sorption/desorption kinetics, mismatched the ratio of dissolved vs. DOC-associated and particle-associated PCBs and substantially underpredicted concentrations, especially for the high chlorinated congeners. The results indicated that some of the common assumptions and paradigms in fate modelling of such hydrophobic compounds should be revisited and models updated.

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* Corresponding author.

E-mail address: antonio.diguardo@uninsubria.it (A. Di Guardo).

1. Introduction

Polychlorinated biphenyls (PCBs) are globally distributed recalcitrant contaminants extensively used in many industrial applications (e.g. dielectric fluids, plasticizer, lubricants, joint sealant) till their restriction in the '80 and regulation with the Stockholm Convention on POPs (Persistent organic pollutants) (IARC, 2016). Even if the primary emissions of POPs have been reduced, environmental reservoirs can act as secondary sources (Nizzetto et al., 2010). Soil is the main reservoir of PCBs in terrestrial ecosystems (Cabrerizo et al., 2011) due to the marked affinity of such hydrophobic chemicals for soil organic matter (Armitage et al., 2006; Cousins et al., 1998). The mobility and bioavailability of PCBs in contaminated soils are generally limited because of their hydrophobicity, the aging processes (e.g. formation of bound residues) and the slow desorption kinetics (Luthy et al., 1997; Pignatello and Xing, 1995; Xing and Pignatello, 1997). However, PCB mobility in soil may be enhanced by the association to dissolved organic carbon (DOC) and mobile organic carbon (OC) coated fine particles/colloid particles (from now on called particles/colloids) (Cousins et al., 1999a; Moeckel et al., 2008) that could play a crucial role in driving the infiltration fluxes of hydrophobic chemicals (Enell et al., 2016; Persson et al., 2008). In the last decade, many environmental fate models have started to implement DOC as mobility enhancer in chemical transport (Ghirardello et al., 2010; Komprda et al., 2009; Lindim et al., 2016; Morselli et al., 2018b; Nizzetto et al., 2016; Terzaghi et al., 2018, 2017), showing its potential in influencing infiltration, runoff, redistribution and fluxes of contaminants towards ground and surface waters. This can be crucial when evaluating the long-term fate of POPs or the bioremediation potential, e.g. in contaminated sites. Recently, Terzaghi et al. (2018) in estimating the time required for PCB natural attenuation using the SoilPlusVeg model (Terzaghi et al., 2017), underlined the need of accurately predict the role of DOC and its dynamics in soil to better estimate the PCB bulk water concentrations and possibly their bioavailability. Several authors (Cousins et al., 1999a, 1999b; McLachlan et al., 2002) suggested that the PCB vertical transport in solid phase is a key feature to perform a correct evaluation of the PCB fluxes (e.g. towards atmosphere). However, they referred to a very surficial soil (up to ~30 cm) and the process was explained as being caused by bioturbation, even though the addition of the transport associated to DOC, colloids/small particles was recommended for future developments (Cousins et al., 1999b).

Furthermore, in order to retain simplicity, many of these models may underestimate the role of several factors regulating the cycle of the soil organic carbon and, as consequence, the chemical mass balance.

Contact time among phases (soil/water, water/DOC), for example, affects both the chemical desorption from soil towards the water (Pignatello and Xing, 1995) and the sorption on DOC (Krop et al., 2001). The time to reach equilibrium partitioning on DOC for hydrophobic chemicals can be brief (minutes) (Krop et al., 2001; Pörschmann et al., 1997; Pörschmann et al., 1998) or relatively long (days) (Haitzer et al., 1999) (Xie et al., 2009) depending on chemical properties and DOC source (Haitzer et al., 1999). Furthermore, DOC quality varies with time (Kamjunke et al., 2017) with potential effects on binding affinity for chemicals (Chin et al., 1997; Chiou et al., 1986, 1987).

Additionally, variables that are generally considered negligible in the variability of the soil/water partitioning processes (e.g. moisture conditions, temperature) can deeply influence the DOC fluxes and particle release, affecting chemical concentrations in mobile phases. Natural occurring wetting-drying cycles, for example, can determine an accumulation of dissolved organic carbon (Huang and Lee, 2001) and promote the mechanical disaggregation of soil particles (Jablonowski et al., 2012). Temperature affects the production and release of DOC from soil due to the mineralization of organic matter, with concentrations generally higher in summer than in winter (Kalbitz et al., 2000).

In the present study, a number of column leaching experiments were performed to investigate the influence of different environmental conditions (soil/water contact time, temperature, saturation) on mobility of PCBs in an aged contaminated soil. Measured concentrations were compared to data predicted by a dynamic air-vegetation-litter-soil model (SoilPlusVeg) (Terzaghi et al., 2017). The goal was to measure the chemical fluxes, focusing on PCB transport associated to DOC and small particles, to gain insights to improve PCBs (as well as other POPs) movement modelling.

2. Materials and methods

2.1. Reagents

Calibrants (PCB 28/31, 52, 101, 138, 153, 180, 209, purity $\geq 99\%$) and internal standards (PCB 30, 155, purity $\geq 99\%$) were purchased from Sigma-Aldrich (St. Louis, MO, U.S.A.), while recovery standards (tetrabromobenzene (TBB), purity 98%; hexabromobenzene (HBB), purity 99%) were purchased from Sigma-Aldrich (St. Louis, MO, U.S.A) and Riedel-de Haën (Seelze, Germany) respectively. Acetone and cyclohexane (pesticide residue grade) were purchased from Fluka Analytical (Sigma-Aldrich, St. Louis, MO, U.S.A). Potassium hydrogen biphthalate (KHP) (purity $\geq 99.95\%$), humic acid (HA, Lot Number: BCBN1711V, Carbon content: 43.12%) and anhydrous sodium sulfate were purchased from Sigma-Aldrich (St. Louis, MO, U.S.A). Florisil (0.150–0.250 mm) was purchased from Merck (Darmstadt, Germany).

2.2. Soil samples

Soil samples were collected from the National Priority Contaminated Site Brescia-Caffaro (Northern Italy). The soil used was obtained from pots which underwent a rhizoremediation experiment, potentially affecting PCB availability. Soil replicates were prepared using the one-dimensional Japanese Slabe Cake (JSC) incremental sampling technique (ITRC, 2012) to reduce concentration variability among replicates. See S1 for more information on site and sampling method.

2.3. Leaching solutions

Two different leaching solutions, tap water (TW) (pH: 6.5; conductivity: 210 $\mu\text{S}/\text{cm}$; DOC concentration: 0.5 mg/L) and humic acid (HA) solution (pH: 6.5; conductivity: 220 $\mu\text{S}/\text{cm}$; DOC concentration: 5.5 mg/L), were used for the experiment. The HA solution was prepared by dissolving Aldrich HA salt in tap water by sonication (Branson sonicator, model 8510, Danbury, CT, U.S.A.) for 1 h and filtering under vacuum through two glass fiber filters with approximate pore size of 1.6 μm and 0.7 μm (GF/A and GF/F, Whatman, Maidstone, England). This apparently low concentration of HA was selected given the higher hydrophobic nature of Aldrich humic acid (Chiou et al., 1987), since the resulting K_{DOC} would be a factor of three higher than those obtained with natural soil humic acid.

2.4. Experimental design

Glass columns (3 replicates for each treatment) (ID: 2.66 cm; length: 50 cm), equipped with a teflon stopcock at the bottom and a 250 mL reservoir at the top, were packed with 2 g of glass wool at the bottom (to prevent soil coarse particle losses) and ~250 g of soil and leached under natural (gravity based) percolation.

The conditions of the experiment varied to account for: 1) increasing contact time (CT), 2) DOC addition in leaching solution (TW vs. HA), 3) temperature (25 °C vs. 15 °C), 4), variation of the saturation conditions (to simulate dry-wetting cycles) (Fig. 1).

The experimental factor "increasing CT" was investigated collecting consecutive fractions of leachate after a variable pre-equilibration time (CT: 2, 5, 7, 48 days) under saturated conditions at 25 °C.

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