Modeling the impact of biota on polychlorinated biphenyls (PCBs) fate and transport in Lake Ontario using a population-based multi-compartment fugacity approach[☆]

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ABSTRACT

Organisms have long been treated as receptors in exposure studies of polychlorinated biphenyls (PCBs) and other persistent organic pollutants (POPs). The influences of environmental pollution on organisms are well recognized. However, the impact of biota on PCB transport in an environmental system has not been considered in sufficient detail. In this study, a population-based multi-compartment fugacity model is developed by reconfiguring the organisms as populated compartments and reconstructing all the exchange processes between the organism compartments and environmental compartments, especially the previously ignored feedback routes from biota to the environment. We evaluate the model performance by simulating the PCB concentration distribution in Lake Ontario using published loading records. The lake system is divided into three environment compartments (air, water, and sediment) and several organism groups according to the dominant local biotic species. The comparison indicates that the simulated results are well-matched by a list of published field measurements from different years. We identify a new process, called Facilitated Biotic Intermedia Transport (FBIT), to describe the enhanced pollution transport that occurs between environmental media and organisms. As the hydrophobicity of PCB congener increases, the organism population exerts greater influence on PCB mass flows. In a high biomass scenario, the model simulation indicates significant FBIT effects and biotic storage effects with hydrophobic PCB congeners, which also lead to significant shifts in systemic contaminant exchange rates between organisms and the environment.

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1. Introduction

Polychlorinated biphenyls (PCBs) are a group of synthetic organic compounds, which is highly accumulative in organisms due to hydrophobicity. (De Laender et al., 2010; Arnot and Gobas, 2004; Jones and de Voogt, 1999; Campfens and Mackay, 1997; Gobas et al., 1988). Mathematical modeling provides an essential basis for estimating the fate and transport of PCBs through an environmental system (Bates et al., 2017; Kelce et al., 1998). Chemical potential and fugacity are two frequently used methods (Mackay, 2001; Campfens and Mackay, 1997). The chemical potential approach utilizes the phase equilibrium thermodynamics, yielding rates of mass diffusion directly proportional to measured concentrations (Neely et al., 1974; Kamaya et al., 1981; Barber, Suérez, and Lassiter, 1988). In 2004, Arnot and Gobas developed a bioaccumulation food web model based on the chemical potential formulation (Arnot and Gobas, 2004), which has been widely used as a standard approach in PCB transport and bioaccumulation studies (McLeod et al., 2015; Selck et al., 2012; De Laender et al., 2010).

However, redox potential is logarithmically related to concentration (non-linear) and can vary significantly due to environmental sensitivity. Therefore it is necessary to establish some standard state at which it has a reference value and separate the environmental sensitivity from contaminant diffusion. (Bates et al., 2017; Mackay et al., 2006; MacLeod et al., 2002; Mackay, 1979). As a

result, the fugacity approach was introduced in the 1980s as a more convenient convention to describe thermodynamic equilibrium. Recent model development also integrates information on multiple and interacting processes on PCBs partitioning and transport in the environment (Hollander et al., 2007). In 2006, Wania created the fugacity-based CoZMo-POP2 model. The model includes 19 environmental compartments and works under dynamic conditions. The model also takes into account seasonal variables and allows for the definition of time-variant emission scenarios (Wania et al., 2006). Furthermore, the fugacity approach has been applied in bioaccumulation and exposure studies for PCBs. A review of bioaccumulation studies using the fugacity approach is provided by Gobas and Morrison, (2000). In 2016, Mackay used the fugacity model to study the processes influencing chemical biomagnification and trophic magnification factors in aquatic ecosystems (Mackay et al., 2016). To allow for further insight, Monte Carlo analysis was introduced to characterize uncertainty under various environmental conditions (De Laender et al., 2010).

Despite these previous improvements, organisms have long been treated as pollution exposure assessment targets, and their biological impacts on pollutant fate and transport are considered only in recent years, regarding specifically the contaminant transport through species migration (Walters and Christensen, 2018; McGill et al., 2017; Krümmel et al., 2003). When establishing the contaminant mass balance within the organisms, the direct exchange processes of PCB mass through biotic compartments are well categorized and formulated. However, the biotic compartments respond more rapidly than the environmental sectors. Depending on the nutrition level, mortality rate, food web, and temperature, the population of a species may shift substantially over relatively short periods. Previous models use growth dilution to represent the volumetric changes of biotic compartments rather than direct contaminant exchanges (Gewurtz et al., 2006; Arnot and Gobas, 2004; Campfens and Mackay, 1997). Since growth dilution expresses reductions in concentration due to an organism's volume expansion, it neither describes the effects of organism population behavior on overall PCB transport in the system nor clarifies the destination of contaminant discharged from the organism.

Highly chlorinated PCB congeners are highly hydrophobic, and the high lipid content of organisms provides an excellent location for storage (Kaur et al., 2012; Kelly et al., 2004; Gobas et al., 1988; Barber et al., 1988). Recent studies have confirmed that the PCB bioaccumulation and storage effects on organism population scales may have a significant impact on PCB transport in an ecosystem (Walters and Christensen, 2018; McGill et al., 2017; Krümmel et al., 2003).

To quantify organism impacts on PCB transport, we extend the existing fugacity approach by integrating organisms with environmental media and using a population-based structure. The new design allows us to evaluate the influence of organism population dynamics on PCB mass flow. Thus, PCB transport among biotic groups not only relies on direct individual exchange processes, such as respiration, food ingestion, metabolism, and so on, but also depends on population features, such as birth, growth, predation, and natural mortality rates (Bates et al., 2017; Bates et al., 2015).

Simulations are based on historical records of PCB loadings to Lake Ontario. Lake Ontario has experienced a long history of industrial activities, resulting in significant PCB pollution (Ashworth, 1987). As one of the earliest areas for PCB pollution research, a substantial amount of field data has been compiled since the 1960s (Rukavina, 1976; Oliver and Niimi, 1988; Soonthornnonda et al., 2011). These measurements provide a valuable basis to explore biological impacts on PCBs, and the model evaluation requires a board set of field data to generate reasonable and credible results.

2. Model description

2.1. Formula for population-based fugacity model

The extended approach is a multi-compartment fugacity model; it is proposed to estimate the mass distribution of PCB congeners simultaneously in environmental compartments and organisms. For lake systems, only three environmental compartments are taken into consideration: air, water, and sediment. The study area is simulated by an idealized but representative space (Table 1).

For mathematical simplicity, internal homogeneity is applied to all compartments. For instance, the air compartment is represented as an equilibrium space with constant density, and aerosols and gaseous air are at equilibrium with aerosols evenly distributed throughout the sub-compartment (MacLeod et al., 2002). We include the air compartment in our model because of its role in atmospheric transport and deposition of PCBs (LimnoTech, 2011; Harner and Bidleman, 1996). Furthermore, a recent study shows a potential inhalation problem of lower-chlorinated congeners in the atmosphere (Grimm et al., 2015). The water compartment includes both the water phase and uniformly suspended particles. However, assuming vertical homogeneity in the sediment compartment is unrealistic, since the PCB content with sediment depth depends on the PCB contamination level during the deposition period. As a computational tradeoff, the current sediment compartment only includes the very top layer of bio-active sediment (~0.1 m) which contains about 10% dry residual mixed with the remaining 90% of the saturated water (in volume fraction). We only use one box to represent each environmental media, because the previous study indicates little variation of PCBs concentration regardless of the number of boxes (Kaur et al., 2012).

According to the fugacity approach, the accumulated PCB mass in compartment i is expressed as:

$$M_i = V_i Z_i f_i \tag{1}$$

where $M_i(mol)$ represents the mass of PCBs accumulated in compartment i; $V_i(m^3)$ is the volume of compartment i; $Z_i\left(\frac{mol}{Pa}\cdot m^3\right)$ is the fugacity capacity of compartment i; $f_i(Pa)$ is the PCB fugacity which represents the PCB congener level in compartment i. Thus, the dynamic change of PCBs in compartment i is estimated as:

$$\frac{dM_i}{dt} = \frac{d(Z_i V_i f_i)}{dt} \tag{2}$$

The formula is transformed through partial difference to become,

$$Z_i V_i \frac{df_i}{dt} = \frac{dM_i}{dt} - V_i f_i \frac{dZ_i}{dt} - Z_i f_i \frac{dV_i}{dt} \tag{3}$$

As shown in formula 3, the change of PCBs fugacity in compartment i has three general contributions: the PCBs mass variation (dM_i/dt), the change in fugacity capacity (dZ_i/dt), and the change in compartment volume (dV_i/dt). All compartments related to PCBs transport involve at least one of these three general processes. Table S1, S2, and S3 in the supplemental material list detailed expressions for all exchange processes, as well as parameter summaries and detailed model configurations (Reavie et al., 2014; Breivik et al., 2007; Pauly and Palomares, 2005; Schwarzenbach et al., 2002; Schneider et al., 2001; Boyd, 1995; Harner and Mackay, 1995; Klute, 1986; Munawar and Munawar, 1986; Mackay et al., 1983; Karickhoff, 1981). The major advance of this model is the explicit inclusion of the organism population,