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Research article

Relationship between geosorbent properties and field-based partition coefficients for pesticides in surface water and sediments of selected agrarian catchments: Implications for risk assessment

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ABSTRACT

Studies on pesticide behavior, adsorption-likelihood, and bioavailability vis-a-vis geosorbent properties and seasons, are critical for understanding pesticide-fate and risks in pesticide-prone environments. We examined the relationship between geosorbent profiles of sediments (percentage sand, silt, clay, organic carbon content) across seasons and occurrence of pesticide residues in surface water and sediment of agricultural catchments at Owan, Ogbesse and Illushi communities of Edo State, Nigeria. Pesticide concentrations were measured monthly in samples of surface water and sediments across the selected sites for 18-months. Pesticide behavior and sorption-likelihoods were examined using partition coefficients K_d (sediment-water coefficient), K_{oc} (sediment-water coefficient normalized for organic carbon) and Log K_{ow} (octane-water coefficient); the relationship between K_d and K_{oc} was also examined. Results of the principal component analysis (PCA) indicated that pesticide levels in sediment and surface water were positively associated with the rainy season, total organic content (TOC), percentage silt and clay in sediment. Field-derived pesticide partition coefficients ($K_d < 100$ and log $K_{oc} < 3$) indicated that pesticide species were largely mobile and less likely to be retained in sediments by adsorption. As such, pesticides irrespective of solubility would end up in surface water, increasing risks for pelagic biota and humans sourcing river water for domestic use. Values of Log K_{ow} indicate that organochlorines including DDT, dieldrin, endrin and heptachlor epoxide portend significant bioaccumulation risks to humans and biota across sites. The relationship between K_d and K_{oc} for each site fitted into a quadratic model; it depicted a biphasic behavior of pesticide adsorption and desorption to sediments revealing that concentration of organic carbon across study sites was a limiting factor determining the extent of pesticide adsorption. This study demonstrates that understanding pesticide mobility using field-based partition coefficients could give a clearer picture of pesticide risks to biota and human populations.

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1. Introduction

Concerns related to the adverse effects of pesticide contamination on the environment and human health has spurred a considerable amount of research into their environmental behavior and fate (Caldas et al., 1999; Hall et al., 2015; Lazartigues et al., 2011; Zhang et al., 2015). According to the International Programme on Chemical Safety IPCS (2003), contaminant fate could be conceptualized as distribution pattern of a contaminant, its derivatives or metabolites across biotic and abiotic interfaces due to transport, partitioning, transformation or degradation. Pesticides can be transported from treated fields into surface water through particle-facilitated transport or runoff water both resulting from the abrasive impact of rainfall on exposed soil (Dortch et al., 2008; Reichenberger et al., 2007). Of particular interest is the particulate matter/geosorbents to which contaminants adsorb to in the environment. These particles are key determinants of contaminant fate because of their ability to vary temporally as well as spatially

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(Gagné et al., 2011). Geosorbents are basically characterized by inorganic fractions i.e. rock particles and minerals of varying size and composition which function as the actual sorbents for contaminants (Tan, 2010). On the basis of size, three fundamental inorganic soil fractions include the coarse fraction (2–0.050 mm) called *sand*, the fine fraction (0.050–0.002 mm) called *silt*, and the very fine fraction (<0.002 mm) referred to as *clay* (Nachtergaele, 2001; Tan, 2010). Documented findings indicate that the mobility and bioavailability of contaminants in soil and sediments are largely determined by the relative occurrence of these inorganic fractions in sediments (Hall et al., 2015). This is because they are ubiquitous, influence the physicochemical properties of soil/sediments and provide highly reactive surfaces/large surface area for contaminant adsorption (Gambrell and Patrick, 1988; Tan, 2010). For instance, sediments with a relatively larger percentage occurrence of smaller particle sizes have been reported to have a higher affinity for contaminants in the environment and portend to have a higher capacity to retain contaminants entering the aquatic system (Che et al., 2003; Horowitz, 1991). Other studies have also demonstrated that contaminants may be bound to various forms of organic matter including living organisms, detritus and coatings on mineral particle, etc. (Davison, 1993; Presley and Trefry, 1980; Tessier et al., 1979). Consequently, the total organic matter in sediment is another measure of sediments' ability to scavenge or retain contaminants (Purushothaman and Chakrapani, 2007; Sharmin et al., 2010). Adsorption i.e. the removal of a compound from bulk solution by its net accumulation at an interface (i.e. surface of a colloidal solid like clay or organic matter), has been unveiled as the key process deciding the ultimate transport, persistence, bioavailability, and ecological risks of pesticides in the environment (Edwards, 2013; Li, 2018; Scheytt et al., 2005). Extensive review of the adsorption-desorption process of pesticides in the environment indicated that it is largely dependent on factors related to soil properties (structure, texture, pH and organic matter content), soil management (farming method i.e. conservation tillage and irrigation method), pesticide properties (water solubility and formulation) and climatic conditions (rainfall and temperature) (Satyanarayana and Wypych, 2004). Other studies have indicated that the magnitude of sorption on geosorbents is modulated by the interaction between soil or sediment properties and structural/chemical characteristics of the contaminants (volatility, solubility, degradability, etc.) (Arias-Estévez et al., 2008). Pesticide mobility between sediment and surface water has been evaluated using sorption coefficients derived from either laboratory or field-based assessments (Landrum et al., 1996). The partition (or distribution) coefficient (K_d) is used for estimating the migration potential of contaminants present in aqueous solutions in contact with surface, subsurface and suspended solids. High K_d values portend higher likelihood for adsorption sediments while those with low K_d values portend weak adsorption due to greater water-solubility and mobility in the soil solution (Landrum et al., 1996). The K_d coefficient has a wide variability in different environments because it does not take into cognizance the organic carbon in sediment but is solely based on the inorganic qualities of sediment (e.g. ionic strength, sorbent surface area, available binding sites, steric hindrance etc. (Landrum et al., 1996; Soma and Soma, 1989). Correlations between organic matter content and the coefficient K_d led to the development of a coefficient normalized for dissolved organic carbon, called K_{oc} (Zacharia, 2011). It provides a relative measure of mobility of compounds in aqueous/sediment environment (Sabljić et al., 1995) and is recognized by industries for risk assessments (Wauchope et al., 2002). Lastly, the octane-water coefficient (Log K_{ow}) is the ratio of concentrations of a dissolved substance in octanol phase compared to its concentrations in the aqueous phase (Finizio et al., 1997; Scheytt et al., 2005). It describes the lipophilicity or hydrophilicity of pesticide species and is notable for its application in ecological risk assessment. The use of partition coefficients in assessing pesticide mobility is built on the premise that the compounds are not actively bio-transformed or degraded (Landrum et al., 1996). A number of studies have documented the incidence of pesticide residues in freshwater matrices of the Nigerian environment (Adeboyejo et al., 2011; Adeyemi et al., 2011; Ezemonye et al., 2015b; Upadhi and Wokoma, 2012; Williams, 2013). This also includes a recent study on Ogbesse river where documented concentrations of pesticide residues ranged from not detected (ND) to 0.43 μg/l in surface water and 0.82 to 2.14 μg/kg dw in sediments (Ezemonye et al., 2015a); they also reported the dominance of organochlorines in both matrices. Similar reports for Owan river with concentration of pesticide residues ranging from ND to 0.43 μg/l for water samples, 0.82 to 2.14 μg/kg/dw for sediment, has also been reported (Ogbeide et al., 2015). Findings from Illushi rice fields indicate that pesticide concentration in water samples ranged from ND to 1.65 μg/l, while the concentration of pesticide residues in sediment ranged from ND to 8.45 μg/kg dw (Ogbeide et al., 2016). Despite these commendable reports, the relationship between pesticide mobility and exposure risks to humans and biota in tropical environments is scarcely emphasized. In this present study, we hypothesize that pesticide bioavailability in water and sediment of aquatic systems in agrarian catchments is significantly attributable to the chemical characteristics of the pesticides being used, variables associated with seasonal changes, and inorganic fractions characterizing geosorbent properties of the receiving aquatic system. Furthermore, this study seeks to demonstrate that site-specific patterns of pesticide bioavailability and partitioning between sediment and surface water may result in different risk scenarios for resident biota and human populations sourcing water from these rivers. Lastly, we seek to describe the relationship between the inorganic capacity of sediments to adsorb pesticides (K_d) and the adsorption due to the presence of organic carbon (K_{oc}) across sites.

2. Materials and methods

2.1. Site description

Edo state of Nigeria is a predominantly agrarian geographical area with a landmass of about 19, 187 km^2 of which about 70% is cultivatable land for crop production. The state has a characteristic tropical climate with a distinct wet and dry season which effectively supports the production of a variety of cash crops and food crops. Sampling sites were chosen from three regions of intense agricultural activities (Fig. 1). Illushi River, located in Illushi town (N: 06° 39′ 59.8''; E: 006° 36′ 34.2″) is a prominent rice-cultivating lowland area. The surrounding areas of the Illushi River are characterized by intensive rice farming. This river takes its path through a predominantly loamy soil type area of Edo state (Fig. 1). The Ogbesse River is located in Ogbesse town (N: 06° 45′ 3.7''; E: 005° 34′ 03.2″) within the forest belt ecological zone. It is prominent for cocoa, plantain and pepper production. The surrounding area of this river is typified by the presence of commercial cocoa farms, which constantly make use of agrochemicals to enhance their production. The Owan River is located in Owan Town (N: 06° 45′ 40″; E: 005° 46′ 07.4″). This town is located in the derived savannah ecological zone of Edo State, Nigeria. Surrounding the river are vast farmlands. The region is also notable for its intensive cocoa and plantain production and other types of crop farming. It is important to note that farm lands at each of these sites have an adjacent all-year round flowing river which was constantly and regularly inundated by run-offs from the surrounding farmlands. Ogbesse and Owan River both take their origin from the sandy clay loam

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