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Environmental monitoring and risk assessment of organophosphate pesticides in aquatic ecosystems of north-west Bangladesh

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HIGHLIGHTS

- Chlorpyrifos, diazinon and quinalphos were more frequent in water and sediment samples.
- The highest concentration of OPPs in water was measured for chlorpyrifos as 9.1 μg/L.
- The highest concentration of OPPs in sediment was measured for diazinon as 51 μg/kg.
- RQs showed higher risks of most OPPs for *Daphnia* than fish and algae.
- The higher-tier PERPEST model refined the risks for invertebrates.

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ABSTRACT

The use of organophosphate pesticides (OPPs) to protect a variety of crops has increased in Bangladesh. OPPs may contaminate surrounding aquatic environments through several routes including spray drift, surface runoff and groundwater leaching. Since it is unknown how much OPP end ups in aquatic environment in Bangladesh, the objectives of the present study were to quantify the residues of ten most commonly used OPPs in water and sediment of water bodies of north-west Bangladesh and to assess their ecological risks for aquatic organisms. The risks of the pesticides in surface water and sediment were assessed using a first-tier risk quotient (RQ) approach. The higher-tier PERPEST model was used to refine the ecological risks of pesticides when RQ indicated a potential risk. Results showed the most frequently detected pesticides that appeared in high concentrations were chlorpyrifos, diazinon and quinalphos in surface water and sediment. The highest concentration of OPPs measured in water was 9.1 μg chlorpyrifos/L (median of 1.95 μg/L), while this was 51 μg diazinon/kg dw (median of 11 μg/kg dw) for sediment. Furthermore, results showed high acute and/or chronic RQs (RQ > 1) in surface water and sediment for chlorpyrifos, diazinon, quinalphos, malathion and fenitrothion. The higher-tier PERPEST model confirmed risks of chlorpyrifos, diazinon, quinalphos and fenitrothion for aquatic insects, micro- and macro-crustaceans which were previously derived by RQ-based risk assessment for aquatic organisms. Furthermore, the results of the PERPEST model also indicated possible indirect effects of these pesticides on algae and macrophytes, community metabolism, rotifers and other macro-invertebrates.

1. Introduction

Agriculture is the single largest sector contributing to the national economy of Bangladesh. About 80% of the country's population resides in rural areas and most of them are somehow, directly or indirectly, employed in agricultural activities (Bhattacharjee

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et al., 2012). A further intensification of agriculture in Bangladesh, however, is needed due to its' ever increasing population (about 157 million people on 147,570 km^2 with 1.05% growth rate; BDP, 2016), as well as land scarcity and food security needs (Dasgupta et al., 2007). Severe agro-climatic conditions (e.g. flash floods, seasonal water scarcity, and salinity intrusion in the coastal belt) pose further challenges to agricultural crop production. To meet the growing demand of food under these harsh conditions, farmers are cultivating high-yielding cultivars of crops to get higher yields (Hasanuzzaman et al., 2017), but most of these cultivars are highly vulnerable to pests and diseases (Ali et al., 2018). Hence, like many other developing countries, pesticides are used extensively in Bangladesh to protect the crops (Shahjahan et al., 2017). The government of Bangladesh also fosters the pesticide use to amplify the agricultural frontiers and to increase output per acre of land (Rahman, 2013). In Bangladesh, the Plant Protection Wing of the Ministry of Agriculture (MoA) controls the pesticide registration process. The Pesticide Technical Advisory Committee grants registration to a brand of pesticide after thorough examination of all reports (Rahman, 2013).

The use of pesticides started in Bangladesh around 1951 and remained negligible until 1960s (Ara et al., 2014), but increased dramatically from 7350 metric tons in 1992 to 45,172 metric tons in 2010 (Rahman, 2013). At present, about 84 pesticide active ingredients belonging to 242 trade names of numerous chemical groups such as organochlorine compounds, organophosphates (including all evaluated ones), carbamates, pyrethroids, neonicotinoids, heterocyclic pesticides, nitro compounds and amides have been registered in Bangladesh and are routinely used in agriculture and household applications (Chowdhury et al., 2012a; Ara et al., 2014). Since organochlorine pesticides have been banned in Bangladesh in 1993 (Matin et al., 1998) due to their high toxicity, persistence, and ability to bioaccumulate and biomagnify in the food chain (Sankararamakrishnan et al., 2005; Sun et al., 2006; Teklu et al., 2016), the agricultural sectors have shifted towards organophosphorous pesticides (OPPs) (Chowdhury et al., 2012b). In Bangladesh, an estimated 35% of the crop-producing area is sprayed with OPPs for a variety of crop protection purposes (Chowdhury et al., 2012a).

OPPs may reach the surrounding aquatic environment through several routes including spray drift, direct runoff, ground water leaching, careless disposal of empty containers and equipment washing (Sankararamakrishnan et al., 2005; Hossain et al., 2015; Sumon et al., 2016, 2017). Due to their bioaccumulation ability, OPPs have been detected in different environmental compartments e.g. surface and ground water (Leong et al., 2007; Rahmanikhah et al., 2010; Bhattacharjee et al., 2012; Chowdhury et al., 2012a; Hossain et al., 2015; Hasanuzzaman et al., 2017), sediment (Xue et al., 2005; Abdel-Halim et al., 2006; Nasrabadi et al., 2011; Kanzari et al., 2012; Masiá et al., 2015), and aquatic organisms (Abdel-Halim et al., 2006; Aktar et al., 2009; Malhat and Nasr, 2011; Yang et al., 2012; Masiá et al., 2015; Otieno et al., 2015) in different parts of the world with concentrations ranging from 0.003 ng chlorpyrifos/L (Rahmanikhah et al., 2010) to 0.8 mg chlorpyrifos/L (Akan et al., 2014) in aqueous matrices and 40 ng diazinon/kg (Masiá et al., 2015) to 4.3 mg diazinon/kg (Akan et al., 2014) in solid matrices. OPPs have raised great concern in the scientific community due to their possible ecological risks to the aquatic ecosystems (Masiá et al., 2015; Wee and Aris, 2017), in particular to arthropod invertebrates (Maltby et al., 2005).

The residues of OPPs in the surface water of different water bodies in Bangladesh including rivers, paddy fields and seasonal ponds, have hardly been monitored (Bhattacharjee et al., 2012; Chowdhury et al., 2012a, 2012b; Uddin et al., 2013; Ara et al., 2014; Hossain et al., 2015; Hasanuzzaman et al., 2017). The few available studies, however, did not quantify the residues of OPPs in sediments from aquatic systems and neither assessed pesticides risks for any of the environmental matrices. Hence, the objectives of the present study were to quantify the residues of ten most commonly used OPPs in water and sediments collected from two different water systems in north-west Bangladesh and to assess the ecological risks to aquatic organisms posed by these residues. This study also aimed to identify further research priorities concerning the risks of pesticides for aquatic ecosystems in Bangladesh.

2. Materials and methods

2.1. The study area

Two types of beels, namely Baitkamari and Pirijpur were selected as study sites. A beel is a deep depression along a river where water remains permanent throughout the year. These beels are located in Islampur upazila area of Jamalpur district in north-west Bangladesh, which lies around 25°04′59.88″N and 89°47′30.12″E (Fig. 1). These beels were chosen because local farmers routinely use a variety of pesticides to protect the crops in their vicinity throughout the year. Rice is the dominant crop in the surrounding of the Baitkamari beel and occasionally water chestnut is grown, whereas around the Pirijpur beel the focus is on vegetable production including eggplant, potato, tomato, cauliflower, cabbage, cucumber, pumpkin, and rice and jute. As it is surrounded by much more intensive agriculture, the Pirijpur beel is hypothesised to be more impacted by the pesticide contamination than the Baitkamari beel. The total area of the Baitkamari and Pirijpur beels in the wet season (June–October) is approximately 55 ha and 3 ha with an average water depth of approximately 5 m and 1 m, respectively. In wet season, the water level of both beels becomes high due to rain and flood water received from nearby Brahmaputra River. In dry season (November–March), however, the area of Baitkamari and Pirijpur beel is reduced to approximately 10 ha and 0.1 ha with an average water depth of about 1.8 m and 0.5 m, respectively. Information on crop cultivation and pesticide use in vicinity of both Baitkamari and Pirijpur beels was collected from agricultural officers of Islampur upazila (Table S1). Since farmers use organophosphate pesticides extensively to protect the crops in the vicinity of both Baitkamari and Pirijpur beels, we selected this groups of pesticides in our study (Table S1).

2.2. Collection and preservation of samples

Both surface water and sediment samples were collected from 15 sampling sites of both the Baitkamari as the Pirijpur beel (Fig. 1). Sampling took place in August 2016 (wet season) and in January 2017 (dry season). Surface water samples (approximately 10 cm below water surface) were collected using the hand grab method in 1 L amber glass bottles, filled up to the seal, leaving no space for air bubbles and stored at 4 °C in dark until analysis (Forrest, 2000). The physico-chemical variables of water including temperature, dissolved oxygen, pH and electrical conductivity were measured in situ using a portable multimeter (Hach, HQ 40d). Turbidity was measured in situ using a Secchi disk. Total alkalinity, ammonia, nitrite, nitrate and phosphate concentrations were measured in the Wet laboratory of the Bangladesh Agricultural University in Mymensingh, according to the methods described in American Public Health Association (APHA, 2005). An Ekman grab (length and width: 15 cm and height: 16.5 cm) was used for sampling the upper sediment (upper 5–10 cm). They were homogenized, sieved (mesh size: 1 mm) and stored in 500 mL plastic bottles at 4 °C in dark until analysis. Organic matter, sediment texture and pH were measured in Soil Science Department of Bangladesh Agricultural