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## Factors influencing organochlorine pesticides distribution in the Brisbane River Estuarine sediment, Australia

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### ABSTRACT

Sediment samples collected from Brisbane River were analysed for organochlorine pesticide residues (OCPs). The factors influencing OCPs distribution in the sediment were investigated using multivariate analytical tools. Thirteen OCPs were detected in the sediment with concentrations ranging between below detection to 83.9 ng/g, and detection frequency > 90%. With the exception of dieldrin, the OCP inputs appear to be historical and may cause adverse ecological impacts. Multi-criteria ranking of the factors influencing the OCPs (except dieldrin) distribution in the sediment revealed that TOC > silt > intensive urban land use > population > seasons. Dieldrin distribution is significantly influenced by season > TOC > silt > intensive urban land use > population. The study helps to prioritise factors required for managing OCPs contamination in sediments and identification of appropriate mitigation measures.

#### 1. Introduction

The large scale use of chemicals in agriculture, manufacturing and homes has led to the widespread distribution of contaminants in the environment. These contaminants pose ecological and public health threats because they are toxic to many species, (not just the target species), persistent in the environment and can bioaccumulate or concentrate in species as they move up the food chain (Gilbert, 2012). Typical examples are organochlorine pesticides (OCPs). OCPs are a group of synthetic chlorinated hydrocarbon pesticides, which break down slowly in the environment and usually accumulate in the fatty tissues of animals. They are inherently toxic and often associated with adverse health effects in non-target organisms (US EPA, 2009). Consequently, public awareness of the potential adverse effects of OCPs on ecological and human health has increased. This has led to the ratification of the Stockholm Convention on persistent organic pollutants (POPs) by many countries (UNEP, 2010).

Global production and use of OCPs were extensive between 1940s–1980s for a wide range of applications including agricultural, domestic and public health (Haynes and Johnson, 2000). Most of them were banned about three decades ago. However, their presence and impacts still lingers due to their high photochemical, biological and chemical resistance to degradation in the environment (Duodu et al., 2016a). In Australia, for example, the use of OCPs were banned or restricted from the early 1980s with a complete phase out in October 2010 (Reid et al., 2013). Nevertheless, residues of OCPs are still detectable in the environment (Duodu et al., 2016a; Reid et al., 2013; Elbagir, 2011; Mueller et al., 2011).

In the environment, OCPs tend to accumulate in soils, sediments and biota because of their hydrophobic character and low solubility in water (Duodu et al., 2016a). Sediments provide habitat and nutrients for aquatic flora and fauna and serves as an archive for pollution indexing because of their long residence time (Duodu et al., 2016b; Liu et al., 2017). Therefore, accumulation of OCPs in sediment could undermine water quality and pose adverse impacts on the aquatic ecosystem.

Studies on OCPs in sediments are predominantly centred on analysing their concentrations in order to assess spatial and temporal distribution and ecological and/or human health risk (Hinojosa-Garro et al., 2016; Zhonghua et al., 2016; Alonso-Hernandez et al., 2014; Lebeuf and Nunes, 2005; Barlas, 2002). In terms of OCPs distribution in sediments, factors such as sediment texture, organic carbon, land use and seasonal variation have been found to exert influence. However, the extent and ranking of these factors in terms of the influence they exert have not been investigated. This has constrained the comprehensive understanding of OCP distribution in sediments and consequently, the effective implementation of sediment pollution mitigation strategies. This paper evaluates the critical factors that affect the

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distribution of OCPs in sediment in terms of: (a) ranking the influence they exert; (b) the variability in concentrations when sediment pollution is affected by similar land uses; and (c) the distribution characteristics of different OCP species. Such information can provide valuable insight into the characteristics of OCP distribution in sediments and the identification of the critical factors which need to be managed to facilitate sediment pollution mitigation.

The area under study was the Brisbane River estuary, the highly urbanised river system in Southeast Queensland (SEQ) of Australia. It drains a catchment area of 13,560 km<sup>2</sup> and supports a population in excess of one million. However, there is no recent comprehensive study on the distribution of OCPs in the sediment of the Brisbane River. especially after the 2011 and 2013 floods. The catchment experiences a sub-tropical climate with distinct wet summer and dry winter seasons (Duodu et al., 2016b). The catchment is categorised by a physio-geographic stratification along a hydrological gradient from lower to upper catchment, varying urbanization and distinct land uses (Tables S1 in Supplementary data). Four primary land use types, namely, natural environment (NA), intensive urban use (IU), agricultural land (AG) and water surfaces (WA) can be identified, with each primary land use consisting of various secondary and tertiary types (Liu et al., 2017). A total of 22 sampling points were selected along a 75 km stretch of the river from the mouth, which can be grouped into four physio-geographical strata, namely, rural (SP1-SP3), residential (SP4-SP12), commercial (SP13-SP18), and industrial (SP19-SP22) sections (Duodu et al., 2017) as shown in Fig. 1 and Table S1 with sampling points coordinates and detailed site description. This enabled the analysis of the spatial distribution of OCPs.

Land use related parameters associated with each sampling point were determined from the percentages of each primary land use that account for the total stormwater draining the area. Detailed breakdown of the different land use data, including primary, secondary and tertiary levels as well as their area extents and percentages, and resident population (POP) for each sampling point can be found in Liu et al. (2017). Data for total organic carbon (TOC) was obtained from Duodu et al. (2017). Sedimentary features (particle sizes) were obtained from Duodu et al. (2016b). Table 1 gives the summary of land use, population data, percentage of total organic carbon (TOC) and sedimentary features for each sampling point.

The sediment samples were collected in the months of June (winter), September (spring), December (summer) 2014, and May (autumn) 2015; thus, spanning both dry and wet seasons. This allowed for the observation of temporal variations in OCP concentrations in sediments. Grab sediment (0–3 cm depth) samples were collected from the 22 sampling locations into pre-cleaned 250 mL glass jars, which were initially stored and transported on ice to the laboratory and stored at -20 °C until further analysis.

Each sample was tested for 18 OCPs specified in EPA method 8081B. including alpha, beta, gamma and delta benzene hexachlorides ( $\alpha$ -BHC,  $\beta$ -BHC,  $\gamma$ -BHC and  $\delta$ -BHC). The other compounds were heptachlor, aldrin, heptachlor-exo-epoxide, alpha-endosulfan, 1, 1-dichloro-2, 2-bis (p-chlorophenyl) ethylene (p,p'-DDE), dieldrin, endrin, beta-endosulfan, p,p'-dichlorodiphenyl dichloroethane (p,p'-DDD), p,p'-dichlorodiphenyltrichloroethane (p,p'-DDT), endrin aldehyde, endosulfan sulfate, endrin ketone and methoxychlor. Pentachloronitrobenzene and 2, 4, 5, 6-Tetrachloro-M-Xylene from Supelco Sigma-Aldrich Pty. Ltd. (NSW, Australia) were used as internal and surrogate standards, respectively. Dionex accelerated solvent extractor (ASE 300) system (Thermo Fisher Scientific Australia Pty Ltd) was employed for the extraction of the OCPs from the sediment. Detection and quantification of OCPs concentrations in the extracts were performed using a Shimadzu Gas Chromatograph and Mass Spectrometer (GC-MS) TQ8030. For quality control and quality assurance purposes, surrogate and internal standards were used during the extraction and testing while randomly selected duplicate samples, field blanks, method blanks and SRM 1941b were analysed along with the samples. Details of the extraction, clean-up and analysis had been published elsewhere (Duodu et al., 2016a).

OCPs concentrations that were below the detection limit were considered to be half the method detection limit. The average concentration of the detected OCPs were evaluated and compared with similar studies undertaken elsewhere as well as the Australian New Zealand sediment quality guidelines. The spatial and temporal distribution patterns across the various physio-geographic strata were then elucidated. Afterwards, multivariate data analysis techniques such as



Fig. 1. Location of sampling points (adapted from Google earth).

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