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Pharmaceuticals residues in selected tropical surface water bodies from Selangor (Malaysia): Occurrence and potential risk assessments

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HIGHLIGHTS

- Pharmaceutical residues have created new issues to human health and environment.
- Ciprofloxacin concentrations were the highest in all the river samples.
- Human risk assessment showed low health risk.
- Ecotoxicological risk assessment indicated moderate risks.

GRAPHICAL ABSTRACT

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ABSTRACT

This study investigated the occurrence of nine pharmaceuticals (amoxicillin, caffeine, chloramphenicol, ciprofloxacin, dexamethasone, diclofenac, nitrofurazone, sulfamethoxazole, and triclosan) and to evaluate potential risks (human health and ecotoxicological) in Lui, Gombak and Selangor (Malaysia) rivers using commercial competitive Enzyme-Linked Immunosorbent Assay (ELISA) kit assays. Physicochemical properties of these rivers showed the surface samples belong to Class II of Malaysian National Water Quality Standards which requires conventional treatment before consumption. All the pharmaceuticals were detected in all three rivers except for triclosan, dexamethasone and diclofenac which were not detected in few of sampling locations in these three rivers. Highest pharmaceutical concentrations were detected in Gombak river in line of being as one of the most polluted rivers in Malaysia. Ciprofloxacin concentrations were detected in all the sampling locations with the highest at 299.88 ng/L. While triclosan, dexamethasone and diclofenac concentrations were not detected in a few of sampling locations in these three rivers. All these nine pharmaceuticals were within the levels reported previously in literature. Pharmaceutical production, wastewater treatment technologies and treated sewage effluent were found as the potential sources which can be related with pharmaceuticals occurrence in surface water samples. Potential human risk assessment showed low health risk except for ciprofloxacin and dexamethasone. Instead, ecotoxicological risk assessment indicated moderate risks were present for these rivers. Nevertheless, results confirmation using instrumental techniques is needed for higher degree of

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specificity. It is crucial to continuously monitor the surface water bodies for pharmaceuticals using a cost-effective prioritisation approach to assess sensitive sub-populations risk.

1. Introduction

Emerging contaminant such as pharmaceuticals has generated new issues and risk implications on water quality (X. Liu et al., 2017; Zhou et al., 2017). Generally, pharmaceuticals released into environment from sewage treatment plant effluent, improper disposal of expired pharmaceutical products, hospital waste, domestic sewage, manufacturing plants waste, runoffs from intensive agricultural operations, and excreta from both human and animals (Ahmed and Kasraian, 2002; Celiz et al., 2009; Jiskra and Hollender, 2013; Khetan and Collins, 2007; Patneedi and Prasadu, 2015; Puckowski et al., 2016; Sirés and Brillas, 2012; Wu et al., 2017). The continuous release of pharmaceutical residues into environment will be ultimately distributed to the aquatic environment and groundwater system via leaching and surface runoff. Pharmaceuticals are known to be stable with low biodegradation and high lipophilicity which tend to bioaccumulate in biological organisms and persistent in the environment. Pharmaceutical residues may also affect physiological functions in biological systems considering that they are potential as endocrine disrupters. Hence, the human health as well as the environment have been at risk mainly due to the pharmaceutical exposure, especially in the aquatic environment that receives treated wastewater effluent which will then be utilised as raw water for drinking water (Gavrilescu et al., 2015; Wu et al., 2015; Zeng et al., 2015).

In relation to this, it should be acknowledged that low concentrations of pharmaceutical in the environment at micro-pollutant level give challenge to identification and quantification (Puckowski et al., 2016). Numerous detection methods such as the instrumental method can be applied in pharmaceutical quantification namely high-performance liquid chromatography (HPLC) and gas chromatography mass spectrometry (GCMS). However, there are several disadvantages to these methods such as high detection limit, high operating costs, high usage of chemicals, chemical waste disposal, and clean up involving large number of sample despite the fact that it has been widely applied in pharmaceutical detection involving environmental samples (Białk-Bielińska et al., 2016; Huo et al., 2007; Mohamed, 2015). On the other hand, non-instrumental methods via immunoassay technique are found to provide an alternative methodology that requires the use of specific combinations of antigen and antibody, which is deemed highly sensitive in pharmaceutical determination involving complex environmental matrixes such as surface and wastewater samples. Commercially available Enzyme-Linked Immunosorbent Assay (ELISA) kits that adopt the immunoassay technique has been developed based on the selectivity and affinity of an antibody for its antigen which need to be performed based on certain validation steps. Currently, ELISA kits are used as a quantitative analysis tool in detecting pollutants that are not detected by other instruments such as LC-MS by broad cross-reactivity of antibodies (Shelver and Smith, 2003; Aga et al., 2005; Bradley et al., 2014). ELISA kits are gaining ground because it involves simple sample preparation steps, reasonable cost, small sample volume usage, quick analysis time and the results are highly correlated with the results obtained from HPLC or GCMS (Fang et al., 2016; Huo et al., 2007). It should be noted that immunoassay technique is faced with several matrix effects, but it is often minimized by dilution or adjusting the medium in standard curve construction (Shelver et al., 2008). Meanwhile, Calisto et al. (2011) emphasized that ELISA kits are suitable for screening purposes in order to identify contaminated areas but instrumental techniques (e.g. GCMS, LC–MS/MS) are required to further analyse samples from specific areas. Previous studies have shown the capability of commercial ELISA kits for pollutant quantification in various environmental samples, namely surface water, wastewater, and groundwater (Amitarani et al., 2002; Huo et al., 2007; Shelver et al., 2008; Calisto et al., 2011; Bahlmann et al., 2012; Bradley et al., 2014).

Consumption of human pharmaceuticals especially to treat and control disease related to obesity is at the rise considering Malaysia has the highest number of overweight and obese people in Asia countries (Chan et al., 2017). In addition, veterinary pharmaceuticals usage to prevent, treat, and control illness as well as to promote animal growth in Malaysia is also increasing (Zakaria, 2017). In most cases, continuous exposure of pharmaceutical residue that are released in the form of excreta from human and animals, sewage treatment plant effluents, and improper pharmaceutical products disposal will eventually end up in aquatic environment and groundwater system. The raw water supply from aquatic environment which contains pharmaceuticals will then be treated and supplied as drinking water to residential areas. Moreover, human can be exposed to pharmaceutical residues in drinking water due to the limited capability of conventional drinking treatment systems. Contrasting to other developing countries in Asia, very limited data is available on environmental presence of pharmaceuticals in Malaysian surface water. Up to the present time, only a few of published studies reported the pharmaceutical concentrations found in surface waters of Malaysia (Al-Odaini et al., 2010; Al-Qaim et al., 2014). Findings from indicated that the presence of pharmaceutical residues in drinking water is due to the incompetency of conventional drinking water treatment plants in Malaysia in removing pharmaceuticals. Most of the published studies, however were more focused on pharmaceuticals by putting a very little emphasis on the risks associated with pharmaceutical residues present in the surface water samples. Apart from the high usage of pharmaceuticals in human health and animal livestock in Malaysia, pharmaceutical residue present in the surface water environment is crucial to be investigated for the purpose of filling the knowledge gap on environmental and human health risks associated with tropical climate of Southeast Asia. Moreover, quantitative findings including environmental and human health risks are vital in order to assess public health exposure, especially in the areas where surface water is used as the source of raw water specifically referring to people who are living downstream.

The present study aims to demonstrate the potential and ability of ELISA kits in pharmaceutical screening for the purpose of providing new insights on the contamination status, particularly involving surface water. Lack of studies in Malaysia clearly reflects the need to investigate the physicochemical properties and pharmaceuticals (amoxicillin, caffeine, chloramphenicol, ciprofloxacin, dexamethasone, diclofenac, nitrofurazone, sulfamethoxazole, and triclosan) occurrence in the surface water samples obtained from Lui, Gombak, and Selangor rivers. Moreover, another purpose of this study was to assess human health and ecotoxicological risks associated with pharmaceutical residues pollution in the river waters investigated in this study. The present study acts as a pioneer in providing the quantitative findings of pharmaceutical pollution through ELISA kit assay utilization, including its risks to human health and environment.

2. Materials and method

2.1. Study area and surface water sampling

Fig. 1 shows the sampling locations involving surface water samples collected from Lui, Selangor, and Gombak rivers. Supplementary 1 also provides detailed information of sampling locations from Lui, Selangor, and Gombak rivers. Lui and Selangor rivers are respectively located in