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Occurrence, fate and transformation of emerging contaminants in water: An overarching review of the field☆

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ABSTRACT

Many of the products and drugs used commonly contain chemical components which may persist through sewage treatment works (STW) and eventually enter the aquatic environment as parent compounds, metabolites, or transformation products. Pharmaceuticals and personal care products (PPCPs) and other emerging contaminants (ECs) have been detected in waters (typically ng/L) as well as more recently bound to sediment and plastic particles (typically ng/g). Despite significant advancement of knowledge since the late 1990s, the fate of these contaminants/transformation products once introduced into the aquatic environment remains relatively unresolved.

This review provides a unique focus on the fate of seven major groups of PPCPs/ECs in the aquatic environment, which is frequently not found in similar works which are often compound or topic-specific and limited in background knowledge. Key findings include: a) some replacements for regulation precluded/banned chemicals may be similarly persistent in the environment as those they replace, b) the adsorption of potentially bioactive chemicals to micro- and nanoplastics is a significant topic with risks to aquatic organisms potentially greater than previously thought, and c) micro-/nanoplastics are likely to remain of significant concern for centuries after regulatory limitations on their use become active due to the slow degradation of macro-plastics into smaller components.

An interdisciplinary perspective on recent advances in the field is presented here in a unique way which highlights both the principle science and direction of research needed to elucidate the fate and transport patterns of aquatic PPCPs/ECs. Unlike similar reviews, which are often topic-specific, here we aim to present an overarching review of the field with focus on the occurrence, transformation and fate of emerging contaminants. Environmental presence of seven major classes of contaminants (analygesics, antibiotics, antineoplastics, beta-blockers, perfluorinated compounds, personal care products and plasticisers), factors affecting contaminant fate, association with plastic micro-/nanoparticles and photochemical transformation are comprehensively evaluated.

1. Introduction and background of the field

Pharmaceuticals, personal care products (PPCPs), and many other synthetic organic compounds have revolutionised modern life and their use is now an indispensable component of a healthy society. Pharmaceutical use in particular is an integral component of establishing and maintaining a healthy population of both humans and livestock. In recent years, PPCPs and other emerging contaminants (ECs) have been detected in sewage effluents, surface- and ground-waters, precipitation, and occasionally drinking waters at trace levels (Ternes, 1998; Mohle et al., 1999; Stackelberg et al., 2004; Thomas and Hilton, 2004; Weigel et al., 2004; Lindqvist et al., 2005; Fent et al., 2006; Jjemba, 2006; Nikolaou et al., 2007; Kasprzyk-Hordern et al., 2008; Benotti and Brownawell, 2009; Mahmoud et al., 2009; Silva et al., 2011; López-Serna et al., 2013; Tijani et al., 2016). Furthermore, these compounds are also shown to be present in suspended solids and river sediment (McClellan and Halden, 2010; Walters et al., 2010; Silva et al., 2011; Wilkinson et al., 2017).

The seemingly ubiquitous environmental presence of

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pharmaceuticals and other so-called 'emerging contaminants', albeit in trace or ultra-trace (below ng/L) concentrations, has become of great concern in recent years (Tijani et al., 2016). Furthermore, the extent of PPCPs and other ECs found in the environment, as well as their often largely unknown transformation products, is virtually limitless as new drugs and replacements for regulatory limited compounds are continually being introduced to market.

Many pharmaceuticals and personal care products contain complex chemical structures, often in combination. The original source of these products can be traced to respective manufacturing plants (e.g., 3303 tonnes of paracetamol were produced in France in 2008). However, after leaving manufacturing plants, their routes of environmental exposure become more complex, less understood, and differ between urban and rural environments (Fig. 1). Every human represents a source of PPCP and other emerging contamination, particularly with increasing use of pharmaceuticals (Table 2). From use of soaps and plastics to prescription and non-prescription pharmaceuticals, the personal care products and pharmaceuticals we use in everyday life will inevitably enter the environment after use and excretion. Un-used pharmaceuticals, disposed of in toilets or drains, as well as partially or un-metabolised medication excretions by humans have been shown to be a significant source of prescription and non-prescription drug contamination, representing up to 90% of input (National Association of Clean Water Agencies, 2012). Olsen et al. (2009) found urine to be the most significant source of PPCP elimination in humans. Pharmaceuticals are often not completely metabolised in the body resulting in some proportion of the parent compound (or conjugated derivative) entering sewage treatment facilities (Table 3) via renal (urine) and biliary (faeces) excretions then introduced into the environment (by STW effluent) after incomplete degradation (Jjemba, 2006; Silva et al., 2011).

Virtually every human and veterinary pharmaceutical, either prescription or non-prescription, has been detected in sewage treatment works (STW) effluent (Table 1) and the aquatic environment at levels typically not exceeding upper ng/L. Personal care product (PCP) contamination often includes residues of surfactants used in detergents and soaps, plasticisers used in product packaging and linings, musk compounds used as fragrances, personal insecticides, disinfectants, among others. So-called other ECs include non-commonly monitored agricultural chemicals (for example many herbicides, pesticides and pharmaceuticals used in animal husbandry) and even residues of recreational drugs and their metabolites (Rosi-Marshall et al., 2015). Recent concern regarding groundwater/aquifer contamination by chemicals used in the hydraulic fracturing of shale (e.g., naphthalene used as a surfactant and tetramethyl ammonium chloride as clay stabiliser) may be considered novel emerging contaminants (Gordalla et al., 2013).

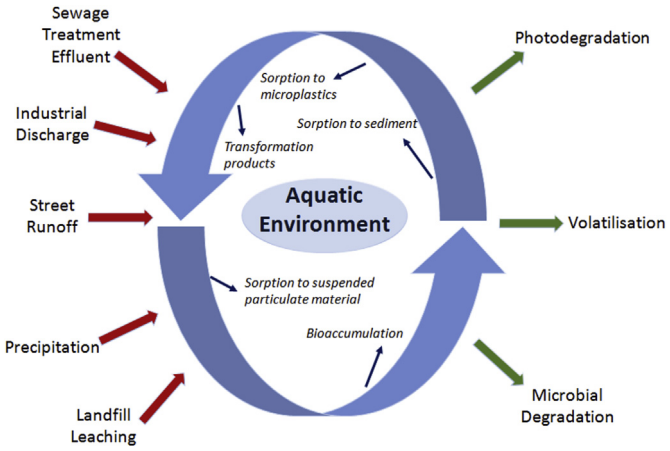

Fig. 1. Sources and fate of PPCP and other ECs in the aquatic environment.

Once in the aquatic environment, other than some well-researched compounds (e.g., BPA and ethinylestradiol), their ultimate fate (i.e., bioaccumulation, spatial distribution, and partition between dissolved and bound to suspended particulate material phases) in aquatic ecosystems is relatively unclear. Most reviews on the study of ECs tend to be limited to a certain chemical (e.g. BPA: Corrales et al., 2015), group of chemicals (e.g. endocrine disrupting compounds: Patisaul and Adewale, 2009; Fisher and Eugster, 2014) or on certain specific aspects of PPCPs/ECs (e.g. removal/degradation during sewage treatment: Luo et al., 2014). This review critically examines the most influential research, encompassing seven major groups of PPCPs and ECs, with a particular focus on occurrence, association with micro-/nanoplastic particles, and bio- and photochemical transformation.

2. The profile of PPCP contamination: what's in the water?

Since the use of synthetic compounds such as pharmaceuticals and personal care products started, these contaminants have been present in the natural and built environments (Jelić et al., 2012). More than 80 compounds and several metabolites have been found in the aquatic environment and some in drinking water, which indicates that not all contaminants are removed during water treatment (Heberer, 2002). While their presence is not a new phenomenon, noticeable attention has been drawn to them within the last decade (Jelić et al., 2012). Arguably much of this novel interest is due to recent research into the ecotoxicological effects on aquatic organisms of other synthetic compounds such as herbicides and pesticides. Product proliferation and ease of access to pharmaceuticals and personal care products have significantly increased the loading of such compounds in both the natural and built environments (National Association of Clean Water Agencies, 2012). The exact combination of compounds in sources of water varies from location to location and concentrations are consistently higher in ground and surface water than in drinking water (Benotti and Brownawell, 2009; National Association of Clean Water Agencies, 2012).

Worldwide, few such compounds have been inventoried or regulated as contaminants (Caliman and Gavrilescu, 2009). Thus, a lack of knowledge exists regarding the occurrence, proliferation, impacts and fate of synthetic pharmaceuticals and personal care products in the environment (Caliman and Gavrilescu, 2009). Benotti and Brownawell (2009) found 21 different synthetic compounds in 19 different municipal water supplies in the United States. In this study, 51 pharmaceuticals were tested for, of which nearly half were detected (Benotti and Brownawell, 2009). The ten most commonly found pharmaceutical contaminants were: atenolol, estrone, carbamazepine, meprobamate, gemfibrozil, naproxen, phenytoin, sulfamethoxazole, trimethoprim, and TCEP (Benotti and Brownawell, 2009). Of the ten most commonly found, median concentrations were less than 10 μg/L (Benotti and Brownawell, 2009). While such concentrations may seem low, some compounds (e.g., ethinylestradiol) have been shown to be endocrine disruptors during critical periods of biological development in fish at concentrations as low as $\leq$1 ng/L (Parrott and Blunt, 2005; Baumann et al., 2014). Thus, particularly in terms of hormonal disruption, concentrations of synthetic contaminants can have biological effects at concentrations detected in environmental samples.

In the United Kingdom, Jones et al. (2002) modelled aquatic