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Review

Occurrence, sources, and fate of pharmaceuticals in aquatic environment and soil

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ABSTRACT

With the rapid economic development, a better living condition leads to longer life expectancy, which increased the total population, in particular the elderly group. It may result in increase in the demand of pharmaceuticals for people in domestic use or in hospital. Although most sewage treatment plants or waste water treatment plantsmet the regulatory requirement, there are still many pharmaceuticals removed incompletely and thus discharged to the environment. Therefore, the pharmaceuticals residue draws the public concern because they might cause adverse effects on the organism even human beings. Recently, many studies have published on the source and occurrence as well as the fate of pharmaceuticals all over the world. This paper summarized and reviewed the recent studies on the sources, occurrence, fate and the effects of the most common pharmaceuticals. Finally, it gave the suggestion and risk management for controlling the pharmaceuticals.

1. Introduction

The term 'chemical of emerging concern' has raised the public attention to the presence in the environment of excessive chemicals used by different kinds of aspect, for instance, industrial and domestic. It is harmful to the humans and the ecosystems because many of them are unregulated or inadequately regulated (Daughton, 2001). One of the particular concerns is 'emerging organic contaminants' (EOCs). It is the term that not only includes the newly discovered compounds but also covers the compounds newly developed in the environment, for instance, pharmaceuticals and personal care products (PPCPs), pesticides, veterinary products, etc (Richardson and Ternes, 2011). One of the EOCs —Pharmaceuticals and Personal care products (PPCPS) has been identified as the chemical of emerging environmental concern recently. It has been widely used as human medicine to treat or prevent diseases, and as veterinary drugs and husbandry growth promoters in agriculture aspects (Halling-Sørensen et al., 1998). The most common pharmaceuticals will be introduced as follows:

1.1. Analgesics/anti-inflammatories

Analgesics are pain-relief drugs that include narcotic analgesics, non-narcotic analgesics, and non-steroidal anti-inflammatory drugs (NSAID). They act in various ways on the peripheral and central nervous systems and are widely used to alleviate the pain present in almost all diseases (Bueno et al., 2012). Common non-narcotic analgesics include acetaminophen and aspirin. Narcotic analgesics include codeine, methadone, morphine and oxycodone. Also, NSAIDs include dcilofenac, fenoprofen, ketoprofen, mefenamic acid, indomethacin, naproxen and ibuprofen (Nicholas-Bateman, 2012).

1.2. Antibiotic

In the late 1930s, natural and synthetic antibiotic were introduced and their usage has increased for human and animal production. It is considered as "pseudo-persistent" which means they enter into the environment continuously and leads to permanent presence. There are more than 10 different classes of antibiotic, namely β-lactams, macrolides, fluoroquinolones, aminoglycosides, sulfonamide and tetracycline. Some of them such as β-lactams (amoxicillin and penicillin) are the most used antibiotic for human therapy (Huang et al., 2001). For veterinary medicine, antibiotic are used to treat disease or to increase feed efficiency and improve growth rates, for example in shrimp hatcheries and cultural ponds (Sarmah et al., 2006).

1.3. Cardiovascular pharmaceuticals(β-blockers/diuretics)

Anti-hypertensive are the drugs used to treat the high blood pressure. There are many kinds of anti-hypertensive, for instance, β-blockers, calcium channel blockers and diuretics.

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1.4. Psycho-stimulants

The most common central nervous system and metabolic stimulant is caffeine. It is commonly found in many drinks and some pharmaceuticals with the aim to reduce the physical fatigue and to restore alertness when drowsiness occurs (Nehlig et al., 1992).Also, caffeine is easily found in surface water (Kolpin et al., 2002).

1.5. Estrogens and hormonal compounds

Hormonal compounds are one of the important classes of pharmaceuticals because of their common use and serious impacts on human and other animals (Santos et al., 2010). The most important natural estrogen includes estriol, estradiol, and estrone which are mainly excreted from human beings. Other important synthetic estrogen such as 17α-ethinylestradiol which is used for contraception by women causes critical effects to the environment such as feminization of male fish, alternation of DNA integrity, immune cell number and ability to breakdown pollutants (Kolpin et al., 2002).

Because of the rise concern of the occurrence of pharmaceuticals in the environment, there are many papers studied the sources and occurrence of them (eg, Heberer, 2002; Eggen et al., 2012; Lapworth et al., 2012; Bueno et al., 2012). Also, there are lots of research focused on the fate of them and the ecotoxicological effects to the organism (eg, Cuklev et al., 2011; Pawlowski et al., 2004; Lyssimachou and Arukwe, 2007; Araujo et al., 2010; Mehinto et al., 2010; Liu et al., 2011; Vannini et al., 2011). This article reviews the sources of the most common pharmaceuticals to surface water, groundwater and the soil from the domestic, hospital, agriculture and industrial discharge. It also defines what are the main sources and the pathway of pharmaceuticals entering to the environment. Thus, the occurrence of selected pharmaceuticals in the water resources and soil zone are going to be evaluated and compared all over the world including Asia, Europe and the America with summarized table. After stating the occurrence in the environment, the fate and the potential effects of them will be discussed. Finally, several suggestion and risk management will be drawn up to warrant the improvement of the sewage treatment plants and raise the public awareness to the concentration of pharmaceuticals.

1.6. Antiepileptic drug

Carbamazepine is one of the antiepileptic drugs that most frequently detected PPCPs in waste water effluent (Zhang et al., 2008). A research showed that the teratogenic effects associated with carbamazepine exposure are due to its matabolites (Amore et al., 1997). Another research showed that carbamazepine is transformed to range of degradation intermediates and produces biological active products through carbamazepine transformative reactions in soil (Li et al., 2013). The incomplete intermediates illustrated that carbamazepine has high risk for transport from soil and contamination of groundwater.

2. Source of pharmaceuticals

The source of the pharmaceuticals can be divided into two ways – point source pollution and diffuse pollution. Point source pollution is a single identifiable source which originates from separate locations and can be calculated in mathematical modeling (Lapworth et al., 2012). For instance, industrial effluent, hospital effluent and sewage treatment plants as well as the septic tank are the major point source to the soil zone and the water resources. On the contrary, diffuse pollution is hard to be identified the discrete location which occurs over board geographical scales (Lapworth et al., 2012). One of the examples is the runoff including agricultural runoff from the animal waste and manure, urban runoff from domestic waste and the leakage from waste treatment systems and plants (Bueno et al., 2012). Compare with point source pollution, diffuse pollution has generally lower environmental loading because it has higher potential for natural attenuation in the soil and subsurface (Murray et al., 2010).

From the below Fig 1, it shows the pathways that how the pharmaceuticals discharged from the sources and then transferred to the receptors. There are six major sources namely, landfill, animal waste, freshwater aquaculture waste, hospital waste, industrial waste and domestic waste which is summarized and simplified by several review literatures (Heberer, 2002; Eggen et al., 2012; Pal et al., 2010; Lapworth et al., 2012). The receptors of pharmaceuticals can be divided as three major bodied in the natural environment, for instance, soil zone, groundwater and surface water.

2.1. Point source

First of all, municipal landfills are the sources of a wide range of compounds with environmental, wildlife and human health concern (Eggen et al., 2012). The waste deposited in the landfill may contain additive that may adversely affect the health of environment. Besides, municipal landfills may generate leachate which contains significant amounts of dissolved organic matter, heavy metal and other contaminants (Li et al., 2009). It is a waste water which is the infiltrating water interacting with fluids, solids and gases within the buried waste (Buszka et al., 2009). According to the study conducted by Buszka et al. (2009), it found that there are several kinds of contaminants such as hormones, pharmaceuticals and fire retardants in the observation wells which are located down gradient from the landfill. Another research by Barnes et al. (2004) in U.S has the similar finding that the well closed to the landfill has high concentration of pharmaceuticals and it is persistent in the groundwater because the landfill generates the organic waste water and leachate that it flows the flow system and infiltrate to the groundwater and is discharged to the surface water.

Secondly, waste water sources are regarded as one of the most important point source of pharmaceuticals to the aquatic environment (Glassmeyer et al., 2005). There are many studies conducted in different countries such as British, Australia, U.S and Spain reporting that the large number ranged from 16 to 54 kinds of pharmaceuticals is found in the waste water (Petrovic et al., 2006; Gros et al., 2006; Bueno et al., 2007; Kasprzyk-Hordern et al., 2008; Batt et al., 2008). From Fig. 1, it shows that waste water is generated by many kinds of aspects such as hospital, aquaculture, industrial and domestic. Most of the waste water is discharged to the sewage treatment plants through the sewage system and some of it enters to the environment directly by releasing the pharmaceuticals from Pharmaceuticals and Personal Care Products (PPCPs) during bathing or swimming (Daughton and Ternes, 1999). Incomplete human metabolism and excretion into waste stream are also the causes of releasing pharmaceuticals to aquatic environment (Kummerer, 2008). Domestic pathway through sewage treatment plants has been recognized as the main route of human pharmaceuticals substances into the aquatic environment (Ternes, 1998). Other sources such as hospital and industrial waste water are also the important sources of the pharmaceuticals entering to the environment, even the seemingly insignificant individual household (Kummerer, 2009). There are up to 16 pharmaceuticals including anti-epileptics and anti-inflammatories are found in the hospital waste water (José Gómez et al., 2006). Some studies have investigated the occurrence of the pharmaceuticals in the effluent and influent of the sewage treatment plants and it found that the removal of the pharmaceuticals is incomplete (Vieno et al., 2007;