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Review article

Occurrence, sources and fate of pharmaceuticals and personal care products in the groundwater: A review

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ABSTRACT

The presence of pharmaceuticals and personal care products (PPCPs) in the aquatic environment may pose potential threat to the ecosystem and human health, hence PPCPs have aroused much concern over the world. The contamination of PPCPs in the groundwater, the main source of drinking water supply in many countries and regions, has been extensively studied in the last decade. This paper reviews the occurrence of frequently detected PPCPs, including antibiotics, anti-inflammatories, lipid-regulators, carbamazepine, caffeine, and N,N-diethyl-m-toluamide in groundwater, with special concern to the progress made over the past three years. Possible emission sources for PPCPs in groundwater, such as wastewater and contaminated surface water, landfills, septic systems, livestock breeding and sewer leakage, are summarized. Besides, adsorption, migration and degradation, the dominant mechanisms in the subsurface transport and fate of PPCPs, are discussed, and the insights into the future study of PPCPs in the groundwater are provided.

Contents

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1. Introduction

For the past decades, a wide range of pharmaceuticals and personal care products (PPCPs) have been repeatedly observed in the aqueous environment worldwide. Among the array are antibiotics, analgesics, steroids, antidepressants, antipyretics, stimulants, antimicrobials, disinfectants, fragrances, cosmetics, and many other chemicals that are widely used on a daily basis for various purposes [1]. Due to their large consumption, PPCPs may enter the aqueous environment directly or indirectly through anthropogenic activities such as sewage discharge, livestock breeding, fertilizing and landfill leachate, resulting in their presence in surface water and groundwater at concentration levels of ng/L to μg/L. It has been shown that continuous exposure to low, subtoxic concentrations of certain PPCPs can cause unexpected consequences and unintended effects on non-target species, and induce undesirable effects on humans and ecosystems [2–5]. Thus their presence in the environment may pose a threat to human and ecological health. Due to insufficient knowledge in terms of toxicity, impacts and behaviors of PPCPs, few are routinely monitored in the environment and many are unregulated [6]. But as their potential for long-term risk to environment is increasingly recognized, the relevant regulations and standards can be expected over the next decades.

To date most of the reviews concerning the occurrence and transformation of PPCPs in water matrices focus mainly on the surface water and wastewater, in which higher concentrations of PPCPs have been identified. Compared to that, reviews regarding the occurrence, sources and fates of PPCPs in groundwater are limited, probably due to the relatively limited studies on this topic. However, as groundwater is an important water resource in many countries and regions and is difficult to remediate once contaminated, it is necessary to expand the knowledge of PPCPs in groundwater. Recently, several studies made countrywide overviews of emerging organic contaminants (EOCs) including PPCPs in the groundwater of Italy [7], Spain [8] and UK [9], providing useful information on the presence, sources, and potential risks to the environment in their countries. Lapworth and co-workers reviewed the occurrence data of EOCs in groundwater worldwide published before 2011, and discussed their sources and pathways [6]. Due to the increasing number of relevant research published after 2011, an updated review of PPCPs in groundwater worldwide is required.

This paper principally outlines the occurrence of several groups of PPCPs that are found ubiquitous in groundwater, including antibiotics, anti-inflammatories, lipid regulators, caffeine, carbamazepine and N,N-diethyl-m-toluamide (DEET), with a special focus on the literature published in the past three years. It also summarizes their possible sources according to their detections in the environment and discusses adsorption and degradation, the dominant mechanisms in the subsurface transport and fate of PPCPs.

2. Occurrence of PPCPs in groundwater

In the past decade, thanks to the progress of analytical techniques, it has become possible and reliable to determine the concentration of PPCPs in groundwater at trace level. The concentrations of PPCPs that are frequently detected in groundwater, especially those reported recently (2012–2014), are shown in Table 1.

2.1. Antibiotics

Antibiotics are extensively used for human and veterinary medicine. Both metabolized and unmetabolized antibiotics secreted through urine and feces have already been detected in wastewater treatment plants and surface water [24]. They are partially degraded in the environment and as a result are likely to accumulate in water bodies. The long-term persistence of antibiotics at low levels can promote the proliferation of antibiotic resistant bacteria in river base flows (the part of streamflow that discharges from groundwater and seeps into streams) and to a certain degree may enhance the drug resistance of microorganisms [25,26]. The presence of antibiotics in groundwater has also aroused much attention around the world recently. A national reconnaissance carried out in the U.S. concerning pharmaceuticals and other organic contaminants in water resources reported the presence of antibiotics in a sampling network of 47 groundwater sites with the detection frequency exceeding 30% [27]. Fick et al. investigated the surface, ground and drinking water possibly polluted by antibiotics and other pharmaceuticals in a region where the bulk drug industry was developed and found high levels of antibiotics, including ciprofloxacin, enoxacin, ofloxacin and trimethoprim in well water samples [28].

Among the various antibiotics, sulfonamides are the most extensively studied and have been found at high concentrations in several studies. Summarized by Lapworth and co-workers, sulfamethoxazole was reported in fifteen different case studies before 2011 [6], and in one case study, an extremely high concentration of sulfonamides (10 μg/L–1 mg/L) was reported in the groundwater down gradient of a landfill site [29]. In recent studies, sulfonamides in the groundwater were detected in five more studies conducted in Switzerland [10], Spain [11,12], USA [15] and China [14]. Although the detected concentrations seemed to be lower than those measured previously [6], the relatively high frequency of detection in some studies indicated that this group of PPCPs should be investigated in future research.

Veterinary antibiotics are frequently reported in the groundwater around breeding facilities and farmland. For instance, Bartelt-Hunt et al. investigated the occurrence of veterinary pharmaceuticals in lagoons and adjacent groundwater at operating swine and beef cattle facilities [30], and sulfonamides like sulfamerazine, sulfamethazine, sulfamethazole and sulfathiazole, along