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Pharmaceuticals and personal care products transference-transformation in aquifer system

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Abstract: Pharmaceuticals and personal care products (PPCPs) are a new kind of contaminant widely existing in the surface water and groundwater environment. In recent years, PPCPs have been received widely attention from many researchers. The migration and transformation of PPCPs are mainly photolysis, biodegradation, adsorption and hydrolysis in aquifer environment. The influencing factors of PPCPs migration include PPCPs' own physical and chemical properties, types and contents of organic matter, pH, lithology, geotechnical structure and the thickness of vadose zone, *etc*. At present, the research of PPCPs in China is still in the primary stage, especially on the contaminant in aquifer system. Therefore, the research in this field needs to be further strengthened.

Keywords: PPCPs; Migration and transformation; Influencing factor; Contaminant

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Introduction

PPCPs refer to pharmaceuticals and personal care products, which was a notion first proposed by Daughton (1999) and widely recognized thereafter. There are a growing number of reports which point out that PPCPs extensively exist in the water environment. Despite the low content (most of them exist in the aquifer environment with the concentrations in the ng/L~μg/L range), PPCPs are closely related to human activities. Due to heavy and frequent use in animal husbandry and human activities, PPCPs seep into the surface water and groundwater in the process of transformation and migration, generating a pseudo-persistent pollutant which threatens human health through drinking water and food (Ternes *et al*. 2002; SUI Qian *et al*. 2010). More than 50 types of PPCPs compounds such as analgesics, antineoplastics, antihypertensives, preservatives and polycyclic aromatics were detected in water environment (Carballa *et al*. 2004). Most PPCPs have strong polarity, bioactivity and opticity with acidic or alkaline functional groups (Snyder *et al*. 2003). PPCPs have been found in many countries and regions, such as the United States (Barnes *et al*. 2008), the United Kingdom (Lapworth *et al*. 2012), Germany (Sacher *et al*. 2001), *etc*.

In recent years, the monitoring data show that the detection rate and concentration of PPCPs are rising all over the world, showing the trend of increasing pollution area and deepening pollution degree. Some countries in Europe and the United States have carried out relevant research on the pollution characteristics and migration and transformation mechanism of PPCPs in the aquifer system. China mainly studies the pollution characteristics of PPCPs in economically developed areas such as Beijing (ZHANG Yan *et al*. 2016), Shanghai (YAO Bo *et al*. 2016) and the Pearl River Delta (LI Kai-yang *et al*. 2016), while the research on PPCPs pollution in groundwater system is less. Using soil column or sandbox as a simulation experimental device to

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study the migration and transformation of PPCPs in groundwater system cannot restore the vadose zone under field conditions, and the undisturbed soil under field conditions is very difficult to collect. If indoor configuration is carried out, the scale effect cannot be well considered. The study mainly focused on the environmental behavior and influencing factors of PPCPs in the transport-transformation photolysis, adsorption, biodegradation and hydrolysis in the aquifer system.

1 Main sources of PPCPs

The main sources of PPCPs in the aquifer system are medicinal products for human and veterinary use and personal care products, as shown in Fig. 1. The medicinal products cannot be fully absorbed, so that the residual parts are excreted via the excretory organ or secretor in the original form or metabolite of the drug. According to the literature, only a small part of the medicinal products are absorbed after being taken and more than a half are excreted with feces and urine. Antibiotics is a typical example (WANG Yong-qiang *et al*. 2019). Drugs for external use and skin care products (shampoo, conditioner, body wash, cosmetics, *etc*.) seep into the domestic sewage when people wash or bathe (WANG Xiao-yan *et al*. 2019). In addition, the leftover and expired medicines and skin care products in bags or bottles as well as medical wastes, if not handled properly, they will wind up in the soil through leakage, and some will infiltrate in soil and water via the sewage treatment plant (WU Man-lin, 2018; Ellis, 2006).

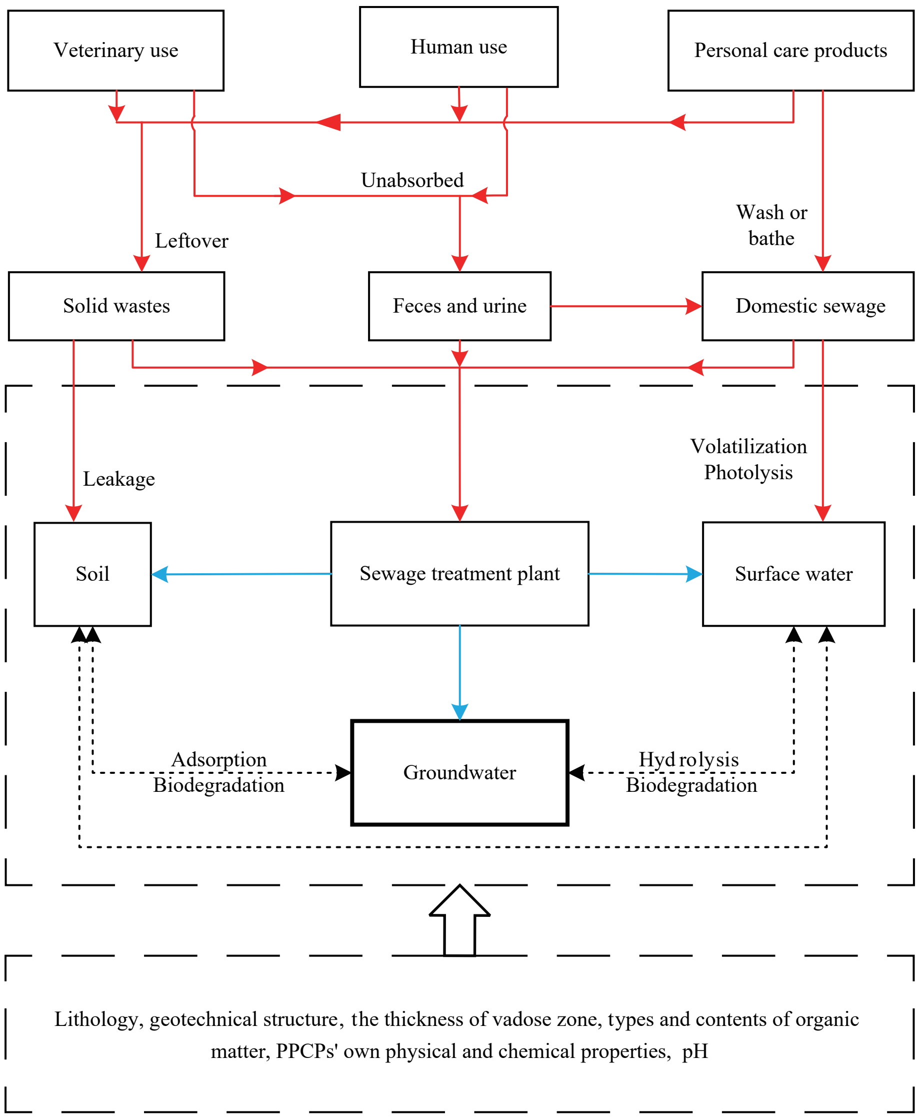

Fig. 1 Transference-transformation of PPCPs in aquatic environment

2 Transference-transformation of PPCPs

Regarding the migration and transformation of PPCPs in the groundwater system, the migration process is mainly affected by the structure and properties of PPCPs, the coefficient of organic carbon adsorption (K_{oc}), the content and type of organic matter, pH in soil, agrotype, vegetation coverage, *etc*. (LU Ying *et al*. 2013; WU Lin *et al*. 2015; ZHU Heng-hua *et al*. 2018). After volatilization and photolysis, the surface residual PPCPs first go into the vadose zone, and then the groundwater by runoff and leaching after a series of complex changes such as adsorption, biodegradation, hydrolysis (ZHANG Xiao-lei *et al.* 2013; SONG Chao, 2016). They may wind up in the drinking water and threaten human health.

2.1 Photolysis

Photolysis is the true decomposition process of PPCPs, which irreversibly changes the reactive molecules. In terms of reaction mechanism, photolysis can be divided into direct photolysis and indirect photolysis (sensitized photolysis). For instance, norfloxacin itself is a photosensitive compound, which will degrade under light (Santos, 2015). GE Lin-ke *et al*. (2009) used two phenicol antibiotics, thiamphenicol and florfenicol, to simulate photolysis in aqueous solution under different light sources. The results showed that humic acids in water can promote the degradation of the two antibiotics under the simulated sunlight ($\lambda > 290$ nm). Lindström *et al.* (2002) did simulation experiments and verified that triclosan, a bactericide, is easy to decompose under sunlight. In August, triclosan's half-life of photodegradation in 47°N area is less than one hour, and its concentration varies with season (lower in summer and higher in winter). WU Yin-bao *et al*. (2006) studied the hydrolysis of enrofloxacin under different pH, microorganism and illumination conditions. The results showed that when pH changed, the microorganism has no significant effect on the degradation of enrofloxacin, but it degrades rapidly under outdoor natural light condition. Andreozzi *et al.* (2003) studied the photodegradation of six representative drugs (carbamazepine, diclofenac, clofibric acid, ofloxacin, sulfamethoxazole and propranolol). The results showed that, in the same conditions, the half-life of carbamazepine and clofibric acid are both 100 days approximately, Sulfamethoxazole, diclofenac, ofloxacin and propranolol have a half-life of 2.4, 5.0, 10.6 and 16.8 days respectively.

2.2 Adsorption

Regarding the migration and transformation of PPCPs (especially hydrophobic PPCPs) in groundwater system, soil adsorption plays a very important role and affects the ecological effect in the process of surface water entering the groundwater through leaching (QIN Qin *et al*. 2019). Amon *et al*. (2008) detected TR (testosterone) and ES (estrogen) in lake sediments at depths of 45 m and 32 m respectively when studying a dairy wastewater lake. The increase of TR, ES, nitrate and chloride contents in the groundwater in monitoring wells nearby indicates that the wastewater lake of the dairy has a significant impact on the groundwater environment.

Scheytt *et al*. (2005) studied the adsorption of carbamazepine (CBZ), diclofenac (DIC) and ibuprofen (IBU) in different sediments, and obtained the soil-water distribution coefficient (K_d) through tests of different batches: K_d of CBZ was 0.21~5.32; K_d of DIC was 0.55~4.66; K_d of IBU was 0.18~1.69. The comparison of the coefficient of organic carbon adsorption (K_{oc}) and related experimental data showed that the calculated data of CBZ is consistent with the experimental data. The calculated DIC and IBU are much higher than the experimental data, which showed that the actual fluidity of DIC and IBU in natural aquifer sediment is higher than the results of correlation equation based on the octanol-water distribution coefficient. Das *et al.* (2004) studied the adsorption capacity of testosterone and its degradation products in soil by conducting soil-column experiment. The research showed that the adsorption capacity of androstenedione, the degradation product of testosterone, is stronger than that of the parent, so that the migration capacity is also weaker, and it is more durable in the environment with the potential risk of secondary pollution.

2.3 Biodegradation

The creature is the most important environmental factor that incurs the decomposition of contaminants. The degradation capability of creatures for different contaminants varies greatly. The difficulty of biodegradation of organic compounds depends first on the properties of the organism itself, and then on the temperature, pH of the environment, structure of organic matters, *etc*. (ZHOU Hai-dong *et al*. 2017).

2.3.1 Microbial degradation

Microbial degradation is the main way for PAHs to disappear in the process of migration and transformation. Although the structure of the compounds in PPCPs is relatively complex, most of the substances can be degraded by the action of microorganisms, and the macromolecular compounds of PPCPs can be transformed into small molecular compounds, finally turning into CO_2 and H_2O. The process is also related to the redox conditions (CHENG Ya-nan *et al*. 2019). In the aerobic environment, oxygen is the main electron acceptor of various biotransformation reactions; in the anaerobic environment, oxygen deficiency, nitrates, sulfates, carbon dioxide and other compounds play the role of electron acceptors. YING Guang-guo *et al*. (2007) conducted simulated experiment of triclocarban and triclosan with the action of microorganisms in soil. The results showed that the half-life of triclocarban and triclosan were 108 days and 18 days respectively under aerobic conditions and the two compounds had existed for 70 days under anaerobic conditions.

2.3.2 Phytodegradation

In the process of migration and transformation, plants can degrade PPCPs through root absorption and root exudates. YANG Yue-qin (2019) studied three plants (Arundo donax var. versicolor, Canna, parasol) and the effect of the vertical flow constructed wetland on buprofen and roxithromycin under non-plant conditions in water. The effects on the removal of roxithromycin and ibuprofen under different seasons, inflow modes and HRT were revealed. Research suggested that different plants had different effects on the removal of ibuprofen and roxithromycin. The best effect on roxithromycin removal was from Arundo donax var.versicolor, reaching 94.06% while Canna had a 69.74% effect on ibuprofen removal. The study found that the presence of plants was more conducive to the removal of ibuprofen and roxithromycin. Rabello studied caffeine, ibuprofen, naproxen, *etc*. The results showed that algal tank may be more effective than constructed wetland on the removal of these drugs (Malta *et al*. 2019).

2.4 Hydrolysis

Hydrolysis is an important way of PPCPs transformation in aquifer environment. Not all PPCPs can be hydrolyzed. Instead, those with amides, carbamates, carboxylates are easy to hydrolyze, for example, tetracycline antibiotics (TCs) (ZHANG Xing-yan *et al*. 2016), β - lactams (AN Bo-yu *et al*. 2019) and sulfonamides (ZHONG Zhen-xing, 2012). The hydrolysis of PPCPs is mainly related to the structure, functional groups and properties of the organic matter, as well as the pH, ionic strength and temperature of the aquifer environment (SONG Chao, 2016; ZHANG Nan, 2012).

3 Influencing factors

PPCPs are closely related to people's life. Despite the low content in the environment, once they diffuse to the air, soil and water, there will be environmental behaviors in the transformation process of photolysis, adsorption, biodegradation and hydrolysis. They may also be affected by physical and chemical properties, pH, type and content of organic matter, lithology, geotechnical structure and thickness of vadose zone. Among them, the type and content of organic matter are the main factors affecting photolysis; lithology, geotechnical structure and the property of vadose zone are the main factors affecting adsorption; the properties of organism and the structural features of organic matter are the main factors affecting biodegradation; the physical and chemical properties of organic matter and pH are the main factors affecting hydrolysis.

3.1 Physical and chemical properties

The physical and chemical properties of PPCPs will affect their degradation rate. WANG Li-ping *et al*. (2009) studied the influence of soil properties on the adsorption of tylosin and sulfamethazine antibiotics. The results showed that the adsorption of soil on PPCPs varied greatly with different types of PPCPs. ZHOU Hai-dong *et al*. (2017) studied the transference-transformation of 13 representative sewage labeling drugs in urban rivers. The results showed that hydrophilic compounds were not easy to be adsorbed by sediments, while the adsorption capacity of hydrophobic compounds depended on their K_{ow} (octanol-water partition coefficient). Hydrophilic compounds transfer to water quickly. The stronger the adsorption capacity of hydrophobic compounds, the slower the transfer will be.

3.2 pH

Different pH of environmental medium leads to different electrical property of PPCPs after ionization, which further causes the change of degradation rate of PPCPs. CHEN Hao *et al*. (2011) studied the migration of sulfamethoxazole (SMX) and ciprofloxacin (CIP) in saturated quartz packed column under different pH and ionic strength, and found that SMX had stronger migration ability than CIP, and was not easy to be adsorbed. When the pH increased to 9.5, 93% CIP were washed out. Andreozzi *et al*. (2004) studied the photolysis and hydrolysis of amoxicillin at different pH conditions. The results showed that the degradation rate of amoxicillin at pH=7.5 was higher than that at pH=5.

3.3 Types and content of organic matter

In the process of migration and transformation, the aquifer environment, which contains some natural dissolved organic matter (DOM), can generate active oxygen atoms under the sunlight and promote the degradation of contaminants (Oliverira *et al*. 2018). BAI Ying (2019) studied the impact of DOM from four different sources on the indirect photolysis of IBU. The results showed that the four kinds of DOM all promoted the indirect photolysis of IBU by producing active radicals. The promotion effects from high to low are as follows: JKHA > SRHA > SRFA > SRNOM. The effects of $^1O_2$ and OH were significant, while ^{3}DOM * basically played no part. Therefore, not all organic matter can promote the degradation of PPCPs. DOM inhibits the oxidation of sulfamethoxazole (SMX) and trimethoprim (TRI), which is conducive to the migration of PPCPs in the aquatic environment (Wenk *et al*. 2011).

Different DOM content in the environment will also affect the migration and transformation ability of PPCPs. It has been proved that the adsorption of soil for sulfa drugs improves with the increase of organic matter content (Sarmah *et al*. 2006).

3.4 Lithology, geotechnical structure and thickness of vadose zone

Different lithology, geotechnical structure and thickness of vadose zone in the environmental medium will lead to different permeability and porosity, which will affect the adsorption and degradation rate of PPCPs in the process of migration. WU Yin-bao *et al*. (2005) studied the adsorption and desorption of enrofloxacin in three groups of soils: Vegetable garden, paddy field and orchard. The results showed that there was a strong adsorption of enrofloxacin in soil, and it was not easy to be washed out. Thus, its migration ability in soil was weak, and it was not easy to pollute groundwater. ZHOU Ai-xia (2015) conducted a migration experiment of sulfonamide antibiotics in the simulation tank, and the results showed that the migration ability of four sulfonamide antibiotics was different in different aquifer media.

3.5 Other factors

There are both natural organic matter and inorganic matter in the environmental medium, and these factors may also affect the degradation rate of PPCPs in the process of migration and transformation. The photolysis rate of IBU is also affected by pH, salinity, NO_3^- and HCO_3^- of the solution, and the degradation process conforms to the pseudo-first order reaction kinetics (BAI Ying *et al*. 2019; LI Fu-hua, 2016). In addition, different plants and microorganisms in the environmental medium, redox conditions and temperatures will affect the degradation rate of PPCPs (Thelusmond *et al*. 2019). Meanwhile, the environmental behaviors in the process of migration and transformation may also influence each other. For example, the adsorption of some PPCPs and their biodegradation are interactive (LI

Jian-zhong, 2013). Some PPCPs can be degraded by microorganisms into products with higher adsorption capacity (CHENG Ya-nan *et al*. 2019), and some products formed by introducing hydroxyl (-OH) into the reactant molecules are more easily degraded by microorganisms.

4 Conclusions

The main sources of PPCPs are medicinal products for veterinary and human uses and personal care products. The degradation of PPCPs in surface water and groundwater environment mainly includes photolysis, biodegradation, adsorption and hydrolysis. The environmental behaviors in these processes may also affect each other. The factors that affect the migration of PPCPs in the surface water and groundwater environment include the physical and chemical properties of PPCPs, the types and contents of organic matter, pH, lithology and geotechnical structure, *etc*. Different organisms (plants and microorganisms) in the environmental medium also lead to different degradation rate of PPCPs which may be subjected to multiple factors concurrently in the process of migration and transformation.

The research on PPCPs in China is still in the primary stage, and the research on the pollution of groundwater system needs significant improvement. First of all, it is in the hope that the analysis and detection techniques will be improved. The work of detecting the types and concentrations of PPCPs residues in the environment will be actively carried out, providing real and reliable data for future research; second, in-depth research of PPCPs in real and complex aquifer system needs to be conducted. Finally, it is of great significance to control and reduce the emission of PPCPs from the source so as to further reduce the harm to groundwater environment and ecology.

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