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## Review

# Sorption of pharmaceuticals and personal care products on soil and soil components: Influencing factors and mechanisms



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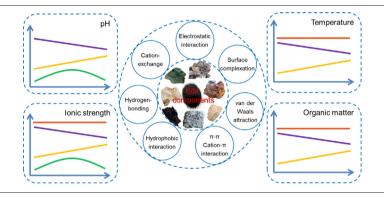
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#### HIGHLIGHTS

## Influencing factors of PPCP sorption to soil and its components were summarized.

- Contrasting trends of PPCP sorption with changes of factors were organized.
- Sorption mechanisms of PPCPs on soil and its components were classified.

#### GRAPHICAL ABSTRACT



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# ABSTRACT

The sorption of pharmaceuticals and personal care products (PPCPs) on soil and soil components makes an important contribution to the fate, migration and bioavailability of PPCPs. Previous reviews have mostly focused on the sorption of PPCPs on single soil components (e.g., minerals and soil organic matter). However, the sorption of PPCPs within the whole soil system has not been systematically analyzed. This paper reviews the recent progress on PPCP sorption on soil and soil components. We have evaluated the sorption of a wide range of PPCPs in research fields that are usually considered in isolation (e.g., humic acids (HAs), montmorillonite, kaolinite, and goethite), and established a bridge between PPCPs and sorbent. The sorption mechanisms of PPCPs, e.g., cation exchange, surface complexation, electrostatic interaction and hydrogen bonding, are discussed and critically evaluated. We also assessed the influence of environmental factors (pH, ionic strength, organic matter and temperature) on sorption. This review summarizes the knowledge of PPCPs sorption on soil gained in recent years, which can provide new strategies for solving the problem of antibiotic pollution.

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#### 1. Introduction

Pharmaceuticals and personal care products (PPCPs) are a cluster of important contaminants, such as various antibiotics, hormones, antiinflammatory drugs and antimicrobial agents (Table S1) (Sören and Thiele-Bruhn, 2003; Tan et al., 2015). With the mass production and application of PPCPs, the environmental pollution caused by them is gradually increasing. Most PPCPs cannot be fully absorbed or metabolized by animal bodies. For example, 30%-90% of ingested antibiotics are excreted into the environment via urine and feces (Liu et al., 2017b; Qin et al., 2016). Residues of PPCPs, derived from municipal wastewater treatment plants and agricultural runoff are frequently detected in surface water, groundwater, soils, and sediments (Boxall et al., 2002; Feng et al., 2016). The widespread use of PPCPs poses potential risks to the environment and human health. For instance, Park and Huwe (2016) have demonstrated that fluoroquinolone antibiotics entering the soil have not only altered the structure of the microbial community, affecting the maintenance of important soil functions, such as nutrient cycling, but also increased the risk of human exposure to antibiotic drugs through contaminated water and food.

The composition of soil is complex, and soils can be divided into three categories in relation to their PPCP sorption capacity: (1) poor sorption capacity, e.g., scree and sand; (2) medium sorption capacity, e.g., iron oxide; and (3) strong sorption capacity, e.g., humic acids (HAs) and montmorillonite (Chang et al., 2016; Scheidegger and Sparks, 1996). Higher clay content and lower sand content in soils can result in a decrease in the mobility of PPCPs, which is attributed to the intense exchange capacity of clay (Tang et al., 2018). The most PPCP-reactive mineral component of soils is clay, and the universal clays in soils are aluminosilicate clays, such as montmorillonite, vermiculite, and kaolinite, whose sorption ability for PPCPs has been widely evaluated (Kocarek et al., 2016). Clay minerals have high specific surface area, cation exchange capacity (CEC) and colloidal dispersion, and have been widely declared as basic sorbents for treating industrial wastewater (Qin et al., 2016). Humic substances have net polyanionic characters over the entire pH range of natural environments. Their net negative charges increase with pH due to the deprotonation of primary carboxyl and phenolic functional groups (Christl et al., 2016; Pignatello, 1999). For example, tetracycline possesses multiple ionizable moieties, in which cations can interact with both low and high proton affinity sites of HA, and zwitterions can interact with high proton affinity sites (Trellu et al., 2016).

Sorption is a crucial process that controls the destiny and mobility of PPCPs in soils (Fig. 1), and in which soil properties such as pH value, organic matter, CEC, and texture play important roles (Qin et al., 2016). Depending on the  $pK_a$  values of PPCPs and soil pH, PPCPs can emerge in nonionic, anionic, cationic or zwitterionic modalities individually or simultaneously (Schaffer and Licha, 2015). Sorption of nonionic molecules is controlled by hydrophobic partitioning to soil organic matter (SOM) via weak van der Waals and electron donor-acceptor interactions (Call et al., 2019). Ionic compounds interact with surface-charged soils through electrostatic interactions such as cation exchange, cation bridging, and complexation. Cationic molecules are mainly adsorbed on negatively charged clay surfaces, and anionic molecules are principally controlled by the positive charges on the solid surface (Call et al., 2019). A better understanding of the factors affecting the sorption of PPCPs is thus essential to better management of pollution in soils (Kah et al., 2017).

Due to the diverse properties of PPCPs and the complex compositions of soils, it is of significance to understand how PPCPs interact with various components of soil, and how environmental factors influence the sorption behaviors, in order to provide essential information on reducing and remediating PPCP contamination in soils. Therefore, this review aims to summarize significant advances in the understanding of the behaviors, mechanisms and influencing factors of sorption of PPCP to soil and components.

## 2. Scope and definitions

## 2.1. Classification and basic properties of PPCPs

PPCPs usually contain a large number of ionic functional groups with multiple acid dissociation constants, so they have obvious ionization tendency (Daughton and Ternes, 1999). PPCPs show different molecular structures and diversity of physicochemical properties (Table S1).

PPCPs can be divided into medical and personal care products, including antibiotics, hormones,  $\beta$ -blockers, cell inhibitors, anti-inflammatory drugs, analgesics and antiepileptics (Daughton and Ternes, 1999). Antibiotics have received special attention because they are the most widely used, which can be divided into several subgroups,

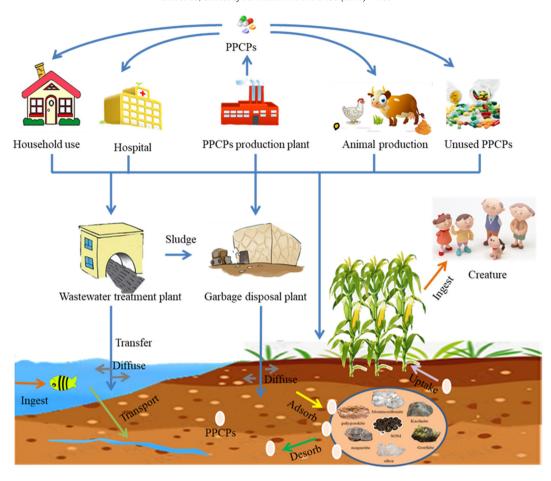


Fig. 1. Potential sources and fate of PPCPs in the environment.

mainly including sulfonamides, fluoroquinolones, tetracyclines, macrolides, and  $\beta$ -lactam. Many of them are amphoteric and ionizable compounds. Synthetic hormones belong to another class of pharmaceutical products, of which 17-estradiol is a representative that affects the ovaries and placenta (Pan et al., 2009). For personal care products, bisphenol A is one of the most commonly detected endocrine compounds in terrestrial environments, which can disrupt human hormone secretion and also affect soil ecosystem functions (Novo et al., 2018). The herbicides and fungicides used in daily agricultural production are also familiar to people (Tian et al., 2016; Zhang et al., 2013a). Monensin is a polyether ion carrier and is a cell inhibitor commonly used in beef and poultry industry to prevent coccidia infection.

## 2.2. Soil and soil components

Soil minerals and organic matter are briefly introduced, since these two components predominantly affect the sorption behavior of PPCPs in soils.

Clay is the most active mineral component in soil. Layered silicate clay minerals can be divided into 1:1 type, 2:1 type and 2:1:1 type according to the stacking sequence and different stacking ratios of silicon wafers and aluminum sheets. Montmorillonite is a typical 2:1 layered silicate structure with permanent negative charges. It has relatively large CEC which is constant with pH value. Its specific surface area can reach 600–800 m² g $^{-1}$ , 80% of which is internal surface area (Simonton et al., 1988). Kaolinite is a typical 1:1 layered aluminosilicate structure. Its mineral surface does not carry permanent negative charge, but its side surface has a typical variable charge surface due to broken bonds, which makes it positively charged under acidic conditions and negatively charged under alkaline conditions (Mcbride, 1978).

Halloysite, which is kaolinite with water molecules between crystal layers, carries permanent negative charges (Joussein et al., 2005). Alchemite and oxides of aluminum and iron are the most important non-layered silicate clay minerals in most soils. The most common iron oxides include goethite and hematite, in addition to magnetite, maghemite, ferric hydroxide or ferrierite (Cornell and Schwertmann, 2004). The oxide colloids of iron and aluminum are amphoteric colloids, and their chargeability changes with the change of environmental pH. Generally, the oxide colloids are positively charged under acidic conditions (pH < 5), while under alkaline conditions, they can be negatively charged (Goldberg et al., 1996).

This paper also deals with other common minerals, such as palygorskite, illite, rectorite. Palygorskite is also called attapulgite, which is a kind of hydrous magnesium aluminum-rich silicate clay mineral with chain layered structure. It carries permanent negative charges and has medium CEC and high surface area (Murray, 2000). Illite is formed by hydration of mica, also known as hydromica. It carries permanent negative charges and has relatively low CEC, generally only  $10\text{--}40~\text{cmol}_\text{c}~\text{kg}^{-1}$  (Jiang et al., 1997). Rectorite is a regular layered clay mineral composed of non-expansive illite and expansive montmorillonite in a ratio of 1:1, so its properties are similar to illite and montmorillonite (Brindley et al., 1982).

Humus is the main source of variable charge in most soils, especially when organic matter content is high. Organic substances in soil, especially the molecular structure of humus, contain various active groups such as carboxyl, phenolic hydroxyl ( $-C_6H_4OH$ ), amino, alcoholic hydroxyl (-OH), enol group (-COH=CH), hydrosulfide group (-SH) and ketone group (-C=O) (Klavins and Purmalis, 2010). The dissociation or protonation of these active groups is the main source of variable charge generated on the surface of humus (Huang, 1995). The negative

charge on the surface mainly comes from the dissociation of carboxyl or hydroxyl functional groups, while the positive charge comes from the protonation of amino groups. However, under normal soil pH conditions, there are few HAs with positive charge, and the vast majority of HAs carry variable negative charge (Gu et al., 2007).

## 3. Influencing factors

The sorption of PPCPs onto soils is affected by multiple factors, including the physicochemical properties of the soil and PPCPs and various environmental conditions (i.e., pH, ionic strength (IS), organic matter, temperature).

# 3.1. Properties of soil components

The sorption behaviors of PPCPs on various soil components or the same components with different structures (e.g., clay) are different. Halloysite has a greater sorption capacity for ciprofloxacin than tourmaline and biotite, which is mainly attributed to its higher specific surface area, void width and pore volume providing a larger number of sorption sites for ciprofloxacin (Duan et al., 2018). The expanded 2:1 clay minerals (e.g., montmorillonite) generally possess greater surface area and CEC than nonexpanded 1:1 clay minerals (e.g., kaolinite), which results in the stronger sorption of enrofloxacin to montmorillonite than to kaolinite. In addition, although two montmorillonites (SWy-2 and STx-1b) have similar structures, there is a significant difference between them: SWy-2 has 12% tetrahedral charge, while STx-1b is zero. The negative charge on SWy-2 tetrahedral sheet enables enrofloxacin to form stronger adsorption complexes on SWy-2 than on STx-1b (Yan et al., 2012).

## 3.2. Properties of PPCPs

The sorption of PPCPs on soil and soil components is also affected by the PPCPs' characteristics, particularly the  $pK_a$  value. Tylosin is a weak base with a p $K_a$  value of 7.1, and it exists as a cation at pH < 7.1. Diatomaceous earth is composed of amorphous silica and metal oxides, and its surface is negatively charged, so the sorption of tylosin on diatomaceous earth mainly relies on electrostatic interaction (Stromer et al., 2018). Nalidixic acid is a weak acid ( $pK_a = 5.95$ ) and exists as zwitterion at pH < 5.95. The highest sorption capacity of nalidixic acid onto montmorillonite with negative charges is observed under acidic conditions. Nalidixic acid is mostly hydrophobic at low pH values, so the hydrophobic effect is the main sorption mechanism in this scenario (Wu et al., 2013b). Most PPCPs with two p $K_a$  values are a cation, zwitterion and anion under acidic, neutral and alkaline conditions, respectively. Their sorption on negatively charged soil components (e.g., clay minerals and SOM) mainly depends on cation exchange or cation bridging. For example, Li et al. (2017b) found that both cations and zwitterions of ofloxacin can be adsorbed to kaolinite through cation exchange. With the increase of pH, the increase of exchange sites on kaolinite leads sorption to reach its maximum at pH 7.0. This phenomenon can also be seen in the sorption of ciprofloxacin on palygorskite (Chang et al., 2016), tetracycline on rectorite (Chang et al., 2009a) and oxytetracycline on nano-hydroxyapatite (Yuan et al., 2019).

#### 3.3. Environmental factors

The effects of environmental factors (i.e., pH, IS, organic matter, temperature) on the sorption of PPCPs to soil and components varied depending on the type of PPCPs and the soil. The distinct results of effects of pH, IS, organic matter, and temperature are summarized in Tables 1, 2, 3 and 4, respectively. We will discuss below the effects of each factor.

# 3.3.1. Effect of pH

## (1) Bell-shaped relationship

Solution acidity generally dominates the existing species of PPCPs, and thus their sorption to soils. Tylosin occurs as a cation and zwitterion under acidic and alkaline conditions, respectively. As a result, the sorption of tylosin on diatomite showed a trend of first strengthening and then weakening with increasing pH, and the highest sorption capacity was observed at a pH of 6.6. This phenomenon can be explained by the fact that under neutral conditions, the sorption of positively charged tylosin to negatively charged diatomaceous earth reaches its peak due to electrostatic interactions, which are weaker under acidic and alkaline conditions (Stromer et al., 2018). A similar bell-shaped effect of pH was also found in the sorption of oxytetracycline on sedimentary soil. When pH was lower than 3.0, the electrostatic repulsion between cationic oxytetracycline and positively charged soil surface resulted in a lower sorption capacity of oxytetracycline. Similarly, at pH > 7.0, electrostatic repulsion between anionic oxytetracycline and negatively charged soil lead to the inhibited sorption of oxytetracycline. The maximum sorption appeared at pH 6.0-7.0, however, this was attributed to the complexation between oxytetracycline and soil particles rather than electrostatic attraction (Feng et al., 2016).

## (2) Decrease in sorption as pH increases

There are two main mechanisms through which increasing pH inhibits the sorption of PPCPs. On one hand, there are changes in charge. For example, the sorption amount of sulfamethoxazole in soil gradually decreases with increasing pH value (Srinivasan et al., 2013). The main reason is that positively charged sulfamethoxazole can be electrostatically attracted by the negatively charged soil surface at low pH. With an increase in pH, the number of positive charges of sulfamethoxazole decreases, and electrostatic repulsion weakened the sorption effect. The sorption of ciprofloxacin and ofloxacin on oil shale also showed this feature. This was consistent with the changes of their charges from positive to negative with the increase of pH, contributing to adverse electrostatic interactions and thus decreased sorption of ciprofloxacin and ofloxacin (Gouza et al., 2017). On the other hand, the change in pH affects the sorption mechanism of PPCPs on soil. For example, the sorption of irbesartan and fexofenadine on soil was mainly driven by cation exchange under acidic conditions (Klement et al., 2018). With the increase of pH, the sorption mechanism changed into cation bridging and surface complexation, but the sorption was greatly reduced due to the repulsion of negative charges on both compounds and soil surface (mainly clay particles). This phenomenon could also be seen in the sorption of flumequine on kaolinite (Li et al., 2019b), in which there were mainly hydrogen-bonding and hydrophobic interactions under acidic conditions, and cationic bridge and surface complexation under alkaline conditions. Electrostatic repulsion also weakened the sorption of flumequine. This phenomenon was also reflected in the sorption of tylosin on montmorillonite. When the pH value of the solution is lower than the  $pK_b$  of tylosin (7.1), the positively charged tylosin is the main species in the solution, and it can be strongly attracted by the negatively charged surface of clay minerals. With an increase in pH value, the sorption mechanism underwent a transition from electrostatic interaction to hydrophobic interaction and then to electrostatic repulsion, and the sorption gradually weakens (Zhang et al., 2013b).

# (3) Increase in sorption as pH increases

Guo et al. (2014) concluded that the sorption of tylosin on goethite was increased with the increasing of pH. Under acidic conditions, tylosin existed in cationic forms, and goethite particles were positively charged. Accordingly, electrostatic repulsion between tylosin and goethite hindered the sorption of tylosin. As pH increased, tylosin became increasingly likely to exist as a neutral species, which had higher affinity than

**Table 1** Effect of pH on the sorption of PPCPs.

| Result               | Sorbate          | Sorbent  | Reference  |
|----------------------|------------------|--|--|
|                      | Norfloxacin      | НА   | (Li et al., 2016; Zhang et al., 2012)  |
|                      | Ciprofloxacin    | Tourmaline, halloysite, biotite, palygorskite, | (Jalil et al., 2015; Liu et al., 2019; Rakshita et al., 2013; Wang et al., 2010a; Zhao                               |
|                      | •                | montmorillonite, magnetite                     | et al., 2018)  |
|                      | Enrofloxacin     | Montmorillonite, kaolinite                     | (Yan et al., 2012)   |
|                      | Ofloxacin        | kaolinite                                      | (Feng et al., 2016; Vieira et al., 2017)   |
|                      | Levofloxacin     | Goethite                                       | (Qin et al., 2016; Wang et al., 2019)  |
|                      | Tylosin          | Diatomaceous earth                             | (Sun et al., 2019)   |
| Bell-shaped          | Spiramycin       | Palygorskite                                   | (Ma et al., 2019)  |
| relationship         | Clarithromycin   | HA   | (Christl et al., 2016)   |
| relationship         | Tetracycline     | Goethite, soil, HA, rectorite, palygorskite    | (Chang et al., 2009a; Chang et al., 2009b; Gu et al., 2007; Jia et al., 2008; Wang et al., 2010b; Zhou et al., 2016) |
|                      | Oxytetracycline  | Sedimentary soil, hydroxyapatite,              | (Feng et al., 2016; Tian et al., 2016; Xu et al., 2016)  |
|                      | Sulfamethoxazole | Soil   | (Kodesova et al., 2015)  |
|                      | Sulfamethazine   | Goethite                                       | (Tri et al., 2018)   |
|                      | Sulfameter       | Soil   | (Zhang et al., 2014)   |
|                      | Nalidixic acid   | Goethite, montmorillonite, kaolinite,          | (Li et al., 2017a; Wang et al., 2017; Wu et al., 2013b)  |
|                      | Niflumic Acid    | Goethite                                       | (Li et al., 2017a; Wang et al., 2017)  |
|                      | Tylosin          | Goethite                                       | (Guo et al., 2014)   |
|                      | Oxytetracycline  | Oil shale                                      | (Park et al., 2016)  |
| Increase in sorption | Sulfamethoxazole | Soil   | (Qin et al., 2016)   |
| as pH increases      | Sulfadiazine     | Soil   | (Qin et al., 2016)   |
|                      | Citalopram       | Soil   | (Call et al., 2019)  |
|                      | Ibuprofen        | Montmorillonite                                | (Martin et al., 2019)  |
|                      | Tylosin          | Montmorillonite, kaolinite, HA,                | (Guo et al., 2015; Zhang et al., 2013b)  |
|                      | Tetracycline     | ferrihydrite, sediments, soil,                 | (Christl et al., 2016; Fernandez-Calvino et al., 2015; Okaikue-Woodi et al., 2018;                                   |
|                      | retracycline     | montmorillonite, HA, Kaolinite                 | Wang and Wang, 2015; Xu and Li, 2010; Zhou et al., 2016)   |
|                      | Oxytetracycline  | Soil   | (Feng et al., 2016)  |
|                      | Sulfamethoxazole | HA, graphite, soil                             | (Ji et al., 2009; Liu et al., 2017a; Srinivasan et al., 2013)  |
|                      | Sulfathiazole    | Montmorillonite, illite, HA, soil              | (Kahle and Stamm, 2007; Kurwadkar et al., 2007; Richter et al., 2009)  |
| Decrease in sorption | Sulfamethazine   | HA, Soil                                       | (Guo et al., 2015; Guo et al., 2016; Kurwadkar et al., 2007; Vithanage et al., 2014; Zhao et al., 2018)              |
| as pH increases      | Sulfadiazine     | Soil, Montmorillonite, kaolinite               | (Xu et al., 2015; Zhang et al., 2019c)   |
|                      | Ciprofloxacin    | HA, montmorillonite, kaolinite, oil shale      | (Liu et al., 2017b; Park et al., 2016; Wu et al., 2010; Zhang et al., 2019c)   |
|                      | Ofloxacin        | Oil shale                                      | (Park et al., 2016)  |
|                      | Flumequine       | Kaolinite                                      | (Feng et al., 2016)  |
|                      | Irbesartan       | Soil   | (Call et al., 2019)  |
|                      | Fexofenadine     | Soil   | (Call et al., 2019)  |
|                      | Pyraoxystrobin   | Soil   | (Yuan et al., 2019)  |

cationic species to goethite. At pH > 9.45, the sorption capacity of tylosin (99.6% of tylosin as neutral species) on goethite reached its peak, in which tylosin can be connected with goethite through  $-NH_2$  and -OH groups.

Martin et al. (2019) found that the sorption of ibuprofen on montmorillonite strongly depends on the pH value of the solution. The sorption efficiency can reach 99.9% at pH 4–9. However, when the pH value fell below 4, the sorption rate dropped significantly from 99.9% to 80.7%.

**Table 2** Effect of ionic strength on the sorption of PPCPs.

| Result   | Sorbate          | Sorbent                                    | References   |
|--|------------------|--|--|
| Bell-shaped relationship                         | Sulfamethoxazole | Soil                                       | (Qin et al., 2016)   |
|  | sulfadiazine     | Soil                                       | (Qin et al., 2016)   |
|  | Sulfapyridine    | Soil                                       | (Srinivasan et al., 2013)                                      |
|  | Sulfamethazine   | HA   | (Zhao et al., 2018)  |
| Increase in sorption as ionic strength increase  | Tetracycline     | Montmorillonite, kaolinite                 | (Kolz et al., 2005; Zhao et al., 2011)                         |
|  | Ciprofloxacin    | Magnetite                                  | (Rakshita et al., 2013)  |
|  | Citalopram       | Soil                                       | (Call et al., 2019)  |
|  | Sulfapyridine    | Montmorillonite                            | (Avisar et al., 2010)  |
|  | Sulfamethoxazole | HA   | (Liu et al., 2017b)  |
|  | Sulfamethazine   | HA, Soil                                   | (Guo et al., 2016; Pavlovic et al., 2014)                      |
|  | Sulfadimethoxine | Soil                                       | (Bialk-Bielinska et al., 2012)                                 |
|  | Sulfaguanidine   | Soil                                       | (Bialk-Bielinska et al., 2012)                                 |
|  | Tetracycline     | Palygorskite, HA, goethite, ferrihydrite   | (Chang et al., 2009b; Christl et al., 2016; Zhou et al., 2016) |
|  | Oxytetracycline  | Nano-hydroxyapatite                        | (Tian et al., 2016)  |
| Decrease in sorption as ionic strength increases | Tylosin          | Kaolinite, montmorillonite, HA<br>Goethite | (Guo et al., 2016; Guo et al., 2014; Shen et al., 2018)        |
|  | Clarithromycin   | HA   | (Christl et al., 2016)   |
|  | Ciprofloxacin    | Sediments, HA                              | (Liu et al., 2017a; Liu et al., 2017b)                         |
|  | Norfloxacin      | HA   | (Li et al., 2016)  |
|  | Ofloxacin        | Kaolinite                                  | (Vieira et al., 2017)  |
|  | Irbesartan       | Soil                                       | (Call et al., 2019)  |
|  | Fexofenadine     | Soil                                       | (Call et al., 2019)  |
|  | Nalidixic Acid   | Goethite                                   | (Li et al., 2017a)   |
| Negliaible effect of ionic atmospath on counting | Sulfapyridine    | Graphite                                   | (Ji et al., 2009)  |
| Negligible effect of ionic strength on sorption  | Niflumic acid    | Goethite                                   | (Li et al., 2017a)   |

**Table 3** Effect of HA on the sorption of PPCPs.

| Result                              | Sorbate          | Sorbent         | References                                    |
|-------------------------------------|------------------|-----------------|---|
|                                     | Ofloxacin        | Kaolinite       | (Feng et al., 2016)                           |
|                                     | Levofloxacin     | Goethite        | (Qin et al., 2016)                            |
|                                     | Sulfamethazine   | Goethite        | (Xu et al., 2017b)                            |
| Increase in sorption as HA increase | Oxytetracycline  | Soil            | (Feng et al., 2016)                           |
| -                                   | Pyraoxystrobin   | Soil            | (Yuan et al., 2019)                           |
|                                     | Naproxen         | Soil            | (Ulmer et al., 2016)                          |
|                                     | Ibuprofen        | Soil            | (Ulmer et al., 2016)                          |
|                                     | Sulfapyridine    | Graphite        | (Ji et al., 2009)                             |
|                                     | Sulfamethoxazole | Soil            | (Narimannejad et al., 2019)                   |
| Decrease in sorption as HA increase | Oxytetracycline  | Soil            | (Feng et al., 2016; Kulshrestha et al., 2004) |
| •                                   | Carbamazepine    | Soil            | (Narimannejad et al., 2019)                   |
|                                     | Pyraoxystrobin   | Soil            | (Yuan et al., 2019)                           |
| Negligible effect of HA on sorption | Enrofloxacin     | Montmorillonite | (Yan et al., 2012)                            |

When the pH value was 1, sorption hardly occurred. The main reason may be that ibuprofen is a weak acid ( $pK_a = 4.5$ ). The sorption of ibuprofen on montmorillonite was mainly through ion exchange between negatively charged ibuprofen and inorganic cations in montmorillonite free sites. Under acidic conditions, this interaction was strongly suppressed.

#### 3.3.2. Effect of ionic strength

#### (1) Bell-shaped relationship

Hu et al. (2019) found that the sorption of sulfamethoxazole and sulfadiazine on silt loam soil increased and then decreased with increasing of IS. The reason for the initial increase was that sulfadiazine and sulfamethoxazole reacted with metal cations and positively charged surfaces to form ternary complexes, a process also called cation bridging, As the concentration of metal ions  $(Ca^{2+}, Mg^{2+})$  in solution increased, there was competitive sorption between the antibiotics and metal cations, resulting in the decreased sorption of antibiotics.

## (2) Decrease in sorption as IS increases

The inhibitory effects of IS on the sorption of PPCPs are mainly divided into the following three categories. (a) The competition between ions and PPCPs for sorption sites. Pavlovic et al. (2017) showed that with the increase of IS, cations (e.g., Ca<sup>2+</sup>) would be electrostatically bonded to the surface of sediments, and inorganic exchangeable cations replaced the hydrogen ions of acidic groups, reducing the number of initial sorption sites. At the same time, the carboxyl and hydroxyl groups of the blocked organic substance cannot be combined with ciprofloxacin molecules through hydrogen bonds, thus reducing sorption. (b) Effects on the electrostatic layer. Bialk-Bielinska et al. (2012) demonstrated that the effect of IS on sorption of sulfadimethoxine on soil was related to the thickness of the electric double layer on the charged

surface. The increase of IS makes the thickness of the electric double layer thinner, resulting in the reduction of negative charges on the soil surface, thus reducing the sorption of positive charges. (c) The changes in conformation of HA. Guo et al. (2016) demonstrated that the increase of IS lead to the shrinkage and reduction of pore size of HA, which resulted in the decrease of the total number of sorption sites, thus weakening the electrostatic interaction between tylosin and HA and reducing the sorption.

## (3) Increase in sorption as IS increases

There are two main mechanisms by which ionic strength promotes the sorption of PPCPs. (a) DLVO theory (named after Derjaguin, Landau, Verwey, and Overbeek, and first proposed to describe the stability of colloidal suspensions). According to DLVO theory, too high IS (10–100 mM) can compress the electric double layer, which can greatly reduce the electrostatic repulsion between HA and sulfamethazine and thus facilitate the sorption to HA (Xu et al., 2016). (b) Cation bridging and salting-out effect. The sorption of sulfamethoxazole on soil increased with the increase of IS, which was attributed to cation bridging. When IS increased,  $Ca^{2+}$  could replace  $H^+$ , and sulfamethoxazole could be complexed with  $Ca^{2+}$ , which was conducive to sorption. Another possibility is that the occurrence of the salting-out effect caused the decrease of solubility of sulfamethoxazole in salt solution, thus precipitating it in the soil. In addition, the increase of IS replaced protons on the soil surface, resulting in a slight decrease of pH value and the transfer of anion sulfamethoxazole to neutral form, which was a more favorable form for sorption to soil particles (Srinivasan et al., 2013).

# (4) Negligible effects of IS on sorption

Some studies also showed that the influence of IS on the sorption of PPCPs in soil or soil medium can be ignored, such as in the case of

**Table 4** Effect of temperature on the sorption of PPCPs.

| Result  | Sorbate            | Sorbent                               | Reference                                   |
|---|--------------------|---------------------------------------|---|
|   | Tetracycline       | Palygorskite, soil                    | (Chang et al., 2009b; Zhang et al., 2013a)  |
|   | Oxytetracycline    | Magnetite                             | (Rakshit et al., 2014)                      |
|   | Sulfamethazine     | HA                                    | (Guo et al., 2015)                          |
| Increase in sorption as temperature increases | Norfloxacin        | HA                                    | (Zhang et al., 2012)                        |
|   | Tylosin            | HA, diatomaceous earth                | (Guo et al., 2015; Sun et al., 2019)        |
|   | Simazine           | Soil                                  | (Zhang et al., 2013a)                       |
|   | Metsulfuron-methyl | Soil                                  | (Zhang et al., 2013a)                       |
|   | Tetracycline       | Layered manganese dioxide birnessite, | (Jiang et al., 2015)                        |
|   | Ciprofloxacin      | Sediments, HA                         | (Liu et al., 2017a; Liu et al., 2017b)      |
| Decrease in sorption as temperature increases | Sulfamethazine     | HA, soil                              | (Vithanage et al., 2014; Zhao et al., 2018) |
|   | Sulfamethoxazole   | HA                                    | (Liu et al., 2017b)                         |
|   | Pyraoxystrobin     | Soil                                  | (Yuan et al., 2019)                         |
| Negligible effect of temperature on sorption  | Spiramycin         | Palygorskite                          | (Ma et al., 2019)                           |

sulfapyridine sorption on graphite (Ji et al., 2009), and niflumic acid sorption on goethite (Xu et al., 2017a).

#### 3.3.3. Effect of organic matter

SOM can promote the sorption of PPCPs to soils in two ways. Firstly, natural organic matter can increase the negative charge on the surface of iron oxides, enhancing electrostatic attraction and thus the sorption of PPCPs with positive charges (e.g., levofloxacin). In addition, the presence of organic matter can improve the stability of iron-oxide colloids and expose more surface areas, promoting the sorption of PPCPs (Qin et al., 2016). Secondly, the formation of compounds can promote sorption. For example, protein substances in dissolved organic matter can be used as bridges to form a compound with goethite and sulfamethazine molecules, thereby improving the sorption of sulfamethazine (Bai et al., 2017).

In contrast, organic matter can also inhibit the sorption of PPCPs. It was reported that the sorption affinity of oxytetracycline on montmorillonite decreased in the presence of a higher concentration of HA (10 mg/L), which was attributed to the fact that more HA molecules were dissolved in solution at high concentrations, and oxytetracycline was desorbed from clay and adsorbed to dissolved HAs (Kulshrestha et al., 2004).

Up to now, SOM has been considered as a critical factor affecting the sorption behavior of hydrophobic organic pollutants. However, for the sorption of ionizable or polar compounds, the effect of organic matter (e.g., HA) might be negligible. For example, with the increase of HA concentration to 5 mg/L, the sorption of enrofloxacin to montmorillonite was unchanged, which might be ascribed to the weaker electrostatic effect of HA than cationic enrofloxacin (Yan et al., 2012).

#### 3.3.4. Effect of temperature

The sorption of PPCPs on soils can be divided into two types: endothermic and exothermic. For endothermic sorption processes, an increase in temperature would accelerate the sorption of PPCPs. In addition, an increase in temperature will reduce the viscosity of the solution and increase the diffusion rate of molecules, helping PPCPs to more easily enter the internal pores through the boundary layer of the sorbent (Guo et al., 2015). This effect was found in the following scenarios: sulfamethazine, tylosin (Guo et al., 2015), and norfloxacin (Zhang et al., 2012)sorption on HA, oxytetracycline (Rakshit et al., 2014) and tetracycline (Zhang et al., 2015) sorption on magnetite, and tetracycline sorption on palygorskite (Chang et al., 2009b).

For exothermic sorption processes, low temperature is more favorable for sorption, e.g., ciprofloxacin (Pavlovic et al., 2017), sulfamethazine (Vithanage et al., 2014), pyraoxystrobin (Liu et al., 2018) sorption on soil, ciprofloxacin (Liu et al., 2017b), sulfamethoxazole (Liu et al., 2017b), and sulfamethazine (Xu et al., 2016) sorption on HA. Additionally, there are cases where temperature did not affect the sorption process, such as spiramycin sorption on palygorskite (Habibi et al., 2018; Ma et al., 2019).

# 4. Sorption mechanisms

When PPCPs interacted with soil, a number of sorption mechanisms may be involved. The sorption mechanisms for various PPCPs that have appeared in the literature are summarized in Table 5 and Fig. 2. The sorption mechanism powerfully hinges on both sorbate (PPCPs) and sorbent (soil and components). Each sorption mechanism will be discussed below.

## 4.1. Electrostatic interaction

# 4.1.1. Cation exchange

Cation exchange is the attraction of a cation to a negatively charged site on the sorbent surface and involves the exchange of one cation for another at the binding site. For the sorption of PPCPs to soils, Vasudevan et al. (2009) found that the sorption potential of ciprofloxacin was controlled by the interaction between the basic structure of ciprofloxacin and the soil, which was mainly ascribed to cation exchange. For the sorption of PPCPs to organic matter, Zhang et al. (2012) stated that in the sorption of norfloxacin on HA extracted from weathered coals, the maximum sorption occurred at pH 6.0, and the interaction between cationic norfloxacin and negatively charged functional groups of HA was dominated by cation exchange. Similarly, Guo et al. (2016) expounded that on the sorption of tylosin and sulfamethazine on HA, pH and IS had significant effects on the adsorption effect, indicating that the main adsorption mechanism was exchange cation. Fourier transform infrared (FTIR) and nuclear magnetic resonance (NMR) analysis also revealed that the amino groups of tylosin and sulfamethazine were the active sorption sites for interaction with HA. For the sorption of PPCPs to minerals, Li et al. (2017b) found that cation exchange contributed to the sorption of cationic ofloxacin to kaolinite. It was reported that the sorption capacity of tetracycline on kaolinite was very high when the pH value was 3.0-6.0, and then decreased with the increasing pH, which indicated that the sorption of tetracycline on kaolinite was realized principally by ion exchange of cation species and complexation of amphoteric ion species (Wang and Wang, 2015; Zhao et al., 2011). The study on the sorption mechanism of tetracycline on clay showed that when the pH was lower than its  $pK_{a1}$  value, tetracycline was in cationic form, and cation exchange was the main mechanism for sorption (Jiang et al., 2015). At pH < 4.0, cationic ciprofloxacin was dominant due to the protonation of the amine group in the piperazine portion, and the surface charges of tourmaline, halloysite and biotite were basically negative, thus, the main sorption mechanism at lower pH values was cation exchange (Duan et al., 2018). Other studies also confirmed the above sorption mechanism (Antilen et al., 2016; Liu et al., 2017c). Cation exchange is also considered as the main sorption mechanism of ciprofloxacin to montmorillonite (Wang et al., 2010a; Wu et al., 2010). Vasudevan et al. (2009) expounded that cationic bridging between the carboxyl group of fluoroquinolones and exchangeable cations associated with negative sites was expected to be achieved by binding the carboxyl group of fluoroquinolones to soil metal oxides and the edge sites of aluminosilicates (Riaz et al., 2018).

Studies have revealed that the sorption of tylosin on kaolinite and montmorillonite was mainly guided by cation exchange; meanwhile, the speciation-dependent sorption model that accounted for the contributions of the cationic forms of tylosin fitted the sorption data well (Guo et al., 2015; Zhang et al., 2013b). In another study, it was found that ion exchange was an important sorption mechanism for electrically charged drugs (i.e., acetaminophen, 17-ethynyl estradiol, nalidixic acid, and norfloxacin) to silica and alumina at environmentally relevant pH values (Lorphensri et al., 2006).

#### 4.1.2. Electrostatic attraction

Electrostatic interaction occurs between a charged sorbent surface and a charged sorbent (Vasudevan et al., 2009). We discussed the possible effects of electrostatic interaction of various PPCPs on soil sorbents. The decrease of solution pH resulted in increased sorption of cationic forms of PPCPs, which indicated that electrostatic force was a favorable sorption mechanism for tylosin and sulfadimidine (Guo et al., 2014; Sassman et al., 2007; Tetzner et al., 2016; Wegst-Uhrich et al., 2014). When sulfonamides were adsorbed to soil, the background ions (Ca<sup>2+</sup> and Mg<sup>2+</sup>) replaced H<sup>+</sup> ions and were complexed with anion species, and sulfonamides thus had a stronger electrostatic affinity to the soil surface (Park et al., 2016). The study by Zhang et al. (2019b) stated that electrostatic interactions between soil particles with negative charges and positively charged sulfonamide were easily generated. Through thermodynamic and FTIR analysis, Hu et al. (2019) proved that the sorption of sulfadiazine and sulfamethoxazole in agricultural soils was a spontaneous physical and chemical sorption process, that was mainly influenced by hydrogen bonds, electrostatic interactions and  $\pi$ - $\pi$  interactions. When ofloxacin was adsorbed on soil, with the

**Table 5**Summary of sorption mechanisms of PPCPs on soil and components reported in the literature.

| Sorption<br>mechanism                     | Sorbate   | Sorbent   | Reference  |
|---|---|---|--|
|   |   | Clay, Clay-humic complexes  | (Carrillo et al., 2016; Chang et al., 2009a; Jia et al., 2008; Jiang et al., 2015; Pils  |
|   | Tetracycline  | Humic materials, layered manganese  | and Laird, 2007)   |
|   | Norfloxacin   | dioxide birnessite, rectorite, quartz sand  | (Li et al. 2016; Perrushi et al. 2015; Thang et al. 2012)  |
|   | Chlortetracycline   | HA, soil<br>Clay, humic substance   | (Li et al., 2016; Peruchi et al., 2015; Zhang et al., 2012)<br>(Pils and Laird, 2007)  |
|   | Ciliorictiacycline  | Clay-humic complexes  | (1 113 dilu Edilu, 2007)   |
|   | Sulfamethazine  | Kaolinite, HA, montmorillonite  | (Teixido et al., 2011; Wang et al., 2015)  |
|   | Sulfamethoxazole  | Soil  | (Berhane et al., 2016; Narimannejad et al., 2019; Wang et al., 2015)   |
|   | Sulfapyridine   | Soil  | (Berhane et al., 2016)   |
|   | Ciprofloxacin   | Soil, humic substances, montmorillonite, halloysite,  | (Liu et al., 2017a; Liu et al., 2019; Pereira Leal et al., 2013; Polesel et al., 2015;   |
|   |   | tourmaline  | Sidhu et al., 2019; Vasudevan et al., 2009; Wang et al., 2010a; Wu et al., 2010;   |
|   | 0 10 11 1   | biotite, palygorskite, river sediments  | Zhao et al., 2018)   |
|   | Sulfadiazine<br>Ofloxacin   | Soil<br>Kaolinite   | (Wang et al., 2015)<br>(Vieira et al., 2017)   |
|   | Enrofloxacin  | Smectite, HAs, soil   | (Gardner and Gunsch, 2017; Yan et al., 2012)   |
|   | Atenolol  | Soil  | (D'Alessio et al., 2019; Narimannejad et al., 2019)  |
|   | Carbamazepine   | Soil  | (D'Alessio et al., 2019)   |
|   | Tylosin   | Soil, montmorillonite   | (Martinez-Hernandez et al., 2016; Sassman et al., 2007; Shen et al., 2018)   |
|   |   | Kaolinite, HA   |  |
|   | Danofloxacin  | Soil  | (Pereira Leal et al., 2013)  |
|   | Oxytetracycline   | Soil, nano-hydroxyapatite, soil organic matter  | (Feng et al., 2016; Jones et al., 2005; Tian et al., 2016)   |
| Cation exchange                           | Sulfamerazine   | Soil  | (Wang et al., 2015)  |
| _   | Sulfadimethoxine<br>Lincomycin  | Soil<br>Smectite  | (Wang et al., 2015)<br>(Wang et al., 2009)   |
|   | β-blocker   | Natural sediment  | (Martinez-Hernandez et al., 2014)  |
|   | atenolol  | Natural Scament   | (Martinez Hernandez et al., 2011)  |
|   | Trimethoprim  | Soil  | (Narimannejad et al., 2019)  |
|   | Tetracycline  | Soil  | (Wang and Wang, 2015)  |
|   | Norfloxacin   | Silica, alumina   | (Lorphensri et al., 2006)  |
|   | Sulfamethazine  | HA  | (Guo et al., 2015)   |
|   | Sulfamethoxazole  | HA  | (dos Santos et al., 2017)  |
|   | Ciprofloxacin   | SOM, HA   | (dos Santos et al., 2017; Goulas et al., 2018)   |
|   | Acetaminophen<br>Tylosin  | Silica, alumina,<br>HA, montmorillonite, kaolinite  | (Lorphensri et al., 2006)<br>(Guo et al., 2015; Zhang et al., 2013b)   |
|   | 17α-ethynyl   | Silica, alumina   | (Lorphensri et al., 2006)  |
|   | estradiol   | Sincu, aranima  | (201) inclusive this 2000)   |
|   | Nalidixic acid  | Silica, alumina   | (Lorphensri et al., 2006)  |
|   | Ofloxacin   | Alumina   | (Goyne et al., 2005)   |
|   | Norfloxacin   | Wetland soil  | (Conkle et al., 2010)  |
|   | Ciprofloxacin   | Wetland soil  | (Conkle et al., 2010)  |
|   | Ofloxacin   | Wetland soil  | (Conkle et al., 2010)  |
|   | Norfloxacin<br>Ciprofloxacin  | Soil<br>Soil  | (Pereira Leal et al., 2013)<br>(Pereira Leal et al., 2013)   |
|   | Danofloxacin  | Soil  | (Pereira Leal et al., 2013)  |
|   |   | Clay, humic substance, clay-humic complexes,  |  |
|   | Tetracycline  | montmorillonite, sediment, soil   | (Pils and Laird, 2007; Zhang et al., 2010)   |
|   | Norfloxacin   | Soil  | (Peruchi et al., 2015)   |
|   |   |   |  |
|   | Chlortetracycline   | Clay, humic substance   | (Pils and Laird, 2007)   |
|   | •   | Clay-humic complexes  |  |
|   | Sulfamethoxazole  | Clay-humic complexes<br>Soil  | (D'Alessio et al., 2019; Srinivasan et al., 2013)  |
|   | •   | Clay-humic complexes<br>Soil<br>Soil, biosolids-amended soil, kaolinite,  | (D'Alessio et al., 2019; Srinivasan et al., 2013)<br>(Chessa et al., 2016; Goulas et al., 2018; Liu et al., 2017a; Liu et al., 2019;   |
|   | Sulfamethoxazole  | Clay-humic complexes<br>Soil<br>Soil, biosolids-amended soil, kaolinite,<br>montmorillonite, tourmaline, biotite, palygorskite,   | (D'Alessio et al., 2019; Srinivasan et al., 2013)<br>(Chessa et al., 2016; Goulas et al., 2018; Liu et al., 2017a; Liu et al., 2019;<br>Pereira Leal et al., 2013; Vasudevan et al., 2009; Zhang et al., 2019c; Zhao et al.  |
|   | Sulfamethoxazole<br>Ciprofloxacin   | Clay-humic complexes Soil Soil, biosolids-amended soil, kaolinite, montmorillonite, tourmaline, biotite, palygorskite, SOM, river sediments   | (D'Alessio et al., 2019; Srinivasan et al., 2013)<br>(Chessa et al., 2016; Goulas et al., 2018; Liu et al., 2017a; Liu et al., 2019;<br>Pereira Leal et al., 2013; Vasudevan et al., 2009; Zhang et al., 2019c; Zhao et al.<br>2018)   |
| Cation bridging                           | Sulfamethoxazole<br>Ciprofloxacin<br>Sulfadiazine   | Clay-humic complexes Soil Soil, biosolids-amended soil, kaolinite, montmorillonite, tourmaline, biotite, palygorskite, SOM, river sediments Agricultural soil   | (D'Alessio et al., 2019; Srinivasan et al., 2013)<br>(Chessa et al., 2016; Goulas et al., 2018; Liu et al., 2017a; Liu et al., 2019;<br>Pereira Leal et al., 2013; Vasudevan et al., 2009; Zhang et al., 2019c; Zhao et al.<br>2018)<br>(Sukul et al., 2008)   |
| Cation bridging                           | Sulfamethoxazole<br>Ciprofloxacin   | Clay-humic complexes Soil Soil, biosolids-amended soil, kaolinite, montmorillonite, tourmaline, biotite, palygorskite, SOM, river sediments   | (D'Alessio et al., 2019; Srinivasan et al., 2013)<br>(Chessa et al., 2016; Goulas et al., 2018; Liu et al., 2017a; Liu et al., 2019;<br>Pereira Leal et al., 2013; Vasudevan et al., 2009; Zhang et al., 2019c; Zhao et al.<br>2018)   |
| Cation bridging                           | Sulfamethoxazole<br>Ciprofloxacin<br>Sulfadiazine<br>Ofloxacin  | Clay-humic complexes Soil Soil, biosolids-amended soil, kaolinite, montmorillonite, tourmaline, biotite, palygorskite, SOM, river sediments Agricultural soil Limestone, alumina, Silica  | (D'Alessio et al., 2019; Srinivasan et al., 2013)<br>(Chessa et al., 2016; Goulas et al., 2018; Liu et al., 2017a; Liu et al., 2019;<br>Pereira Leal et al., 2013; Vasudevan et al., 2009; Zhang et al., 2019c; Zhao et al.<br>2018)<br>(Sukul et al., 2008)<br>(Goyne et al., 2005; Wu et al., 2014)  |
| Cation bridging                           | Sulfamethoxazole<br>Ciprofloxacin<br>Sulfadiazine<br>Ofloxacin<br>Enrofloxacin<br>Danofloxacin<br>Oxytetracycline   | Clay-humic complexes Soil Soil, biosolids-amended soil, kaolinite, montmorillonite, tourmaline, biotite, palygorskite, SOM, river sediments Agricultural soil Limestone, alumina, Silica HA, soil Soil Soil   | (D'Alessio et al., 2019; Srinivasan et al., 2013) (Chessa et al., 2016; Goulas et al., 2018; Liu et al., 2017a; Liu et al., 2019; Pereira Leal et al., 2013; Vasudevan et al., 2009; Zhang et al., 2019c; Zhao et al. 2018) (Sukul et al., 2008) (Goyne et al., 2005; Wu et al., 2014) (Gardner and Gunsch, 2017) (Pereira Leal et al., 2013) (Jones et al., 2005)   |
| Cation bridging                           | Sulfamethoxazole<br>Ciprofloxacin<br>Sulfadiazine<br>Ofloxacin<br>Enrofloxacin<br>Danofloxacin<br>Oxytetracycline<br>Sulfadimethoxine   | Clay-humic complexes Soil Soil, biosolids-amended soil, kaolinite, montmorillonite, tourmaline, biotite, palygorskite, SOM, river sediments Agricultural soil Limestone, alumina, Silica HA, soil Soil Soil Limestone   | (D'Alessio et al., 2019; Srinivasan et al., 2013) (Chessa et al., 2016; Goulas et al., 2018; Liu et al., 2017a; Liu et al., 2019; Pereira Leal et al., 2013; Vasudevan et al., 2009; Zhang et al., 2019c; Zhao et al. 2018) (Sukul et al., 2008) (Goyne et al., 2005; Wu et al., 2014) (Gardner and Gunsch, 2017) (Pereira Leal et al., 2013) (Jones et al., 2005) (Tetzner et al., 2016)  |
| Cation bridging                           | Sulfamethoxazole<br>Ciprofloxacin  Sulfadiazine Ofloxacin Enrofloxacin Danofloxacin Oxytetracycline Sulfadimethoxine Sulfacetamide  | Clay-humic complexes Soil Soil, biosolids-amended soil, kaolinite, montmorillonite, tourmaline, biotite, palygorskite, SOM, river sediments Agricultural soil Limestone, alumina, Silica HA, soil Soil Soil Limestone Limestone   | (D'Alessio et al., 2019; Srinivasan et al., 2013) (Chessa et al., 2016; Goulas et al., 2018; Liu et al., 2017a; Liu et al., 2019; Pereira Leal et al., 2013; Vasudevan et al., 2009; Zhang et al., 2019c; Zhao et al. 2018) (Sukul et al., 2008) (Goyne et al., 2005; Wu et al., 2014) (Gardner and Gunsch, 2017) (Pereira Leal et al., 2013) (Jones et al., 2005) (Tetzner et al., 2016)  |
| Cation bridging                           | Sulfamethoxazole<br>Ciprofloxacin  Sulfadiazine Ofloxacin Enrofloxacin Danofloxacin Oxytetracycline Sulfadimethoxine Sulfacetamide Tetracycline   | Clay-humic complexes Soil Soil, biosolids-amended soil, kaolinite, montmorillonite, tourmaline, biotite, palygorskite, SOM, river sediments Agricultural soil Limestone, alumina, Silica HA, soil Soil Soil Limestone Limestone Montmorillonite   | (D'Alessio et al., 2019; Srinivasan et al., 2013) (Chessa et al., 2016; Goulas et al., 2018; Liu et al., 2017a; Liu et al., 2019; Pereira Leal et al., 2013; Vasudevan et al., 2009; Zhang et al., 2019c; Zhao et al. 2018) (Sukul et al., 2008) (Goyne et al., 2005; Wu et al., 2014) (Gardner and Gunsch, 2017) (Pereira Leal et al., 2013) (Jones et al., 2005) (Tetzner et al., 2016) (Tetzner et al., 2016) (Okaikue-Woodi et al., 2018)  |
| Cation bridging                           | Sulfamethoxazole<br>Ciprofloxacin  Sulfadiazine Ofloxacin Enrofloxacin Danofloxacin Oxytetracycline Sulfadimethoxine Sulfacetamide Tetracycline Sulfathiazole   | Clay-humic complexes Soil Soil, biosolids-amended soil, kaolinite, montmorillonite, tourmaline, biotite, palygorskite, SOM, river sediments Agricultural soil Limestone, alumina, Silica HA, soil Soil Soil Limestone Limestone Limestone Montmorillonite SOM   | (D'Alessio et al., 2019; Srinivasan et al., 2013) (Chessa et al., 2016; Goulas et al., 2018; Liu et al., 2017a; Liu et al., 2019; Pereira Leal et al., 2013; Vasudevan et al., 2009; Zhang et al., 2019c; Zhao et al. 2018) (Sukul et al., 2008) (Goyne et al., 2005; Wu et al., 2014) (Gardner and Gunsch, 2017) (Pereira Leal et al., 2013) (Jones et al., 2005) (Tetzner et al., 2016) (Tetzner et al., 2016) (Okaikue-Woodi et al., 2018) (Richter et al., 2009)   |
| Cation bridging                           | Sulfamethoxazole<br>Ciprofloxacin  Sulfadiazine Ofloxacin Enrofloxacin Danofloxacin Oxytetracycline Sulfadimethoxine Sulfacetamide Tetracycline   | Clay-humic complexes Soil Soil, biosolids-amended soil, kaolinite, montmorillonite, tourmaline, biotite, palygorskite, SOM, river sediments Agricultural soil Limestone, alumina, Silica HA, soil Soil Soil Limestone Limestone Montmorillonite   | (D'Alessio et al., 2019; Srinivasan et al., 2013) (Chessa et al., 2016; Goulas et al., 2018; Liu et al., 2017a; Liu et al., 2019; Pereira Leal et al., 2013; Vasudevan et al., 2009; Zhang et al., 2019c; Zhao et al. 2018) (Sukul et al., 2008) (Goyne et al., 2005; Wu et al., 2014) (Gardner and Gunsch, 2017) (Pereira Leal et al., 2013) (Jones et al., 2005) (Tetzner et al., 2016) (Tetzner et al., 2016) (Okaikue-Woodi et al., 2018)  |
| Cation bridging                           | Sulfadiazine Ofloxacin Enrofloxacin Danofloxacin Oxytetracycline Sulfadimethoxine Sulfacetamide Tetracycline Sulfathiazole Tetracycline Ciprofloxacin   | Clay-humic complexes Soil Soil, biosolids-amended soil, kaolinite, montmorillonite, tourmaline, biotite, palygorskite, SOM, river sediments Agricultural soil Limestone, alumina, Silica HA, soil Soil Soil Limestone Limestone Limestone Montmorillonite SOM Kaolinite, soil   | (D'Alessio et al., 2019; Srinivasan et al., 2013) (Chessa et al., 2016; Goulas et al., 2018; Liu et al., 2017a; Liu et al., 2019; Pereira Leal et al., 2013; Vasudevan et al., 2009; Zhang et al., 2019c; Zhao et al. 2018) (Sukul et al., 2008) (Goyne et al., 2005; Wu et al., 2014) (Gardner and Gunsch, 2017) (Pereira Leal et al., 2013) (Jones et al., 2005) (Tetzner et al., 2016) (Tetzner et al., 2016) (Okaikue-Woodi et al., 2018) (Richter et al., 2009) (Wang and Wang, 2015; Zhao et al., 2011) (Chessa et al., 2016)  |
| Cation bridging                           | Sulfamethoxazole<br>Ciprofloxacin  Sulfadiazine Ofloxacin Enrofloxacin Danofloxacin Oxytetracycline Sulfadimethoxine Sulfacetamide Tetracycline Sulfathiazole Tetracycline  | Clay-humic complexes Soil Soil, biosolids-amended soil, kaolinite, montmorillonite, tourmaline, biotite, palygorskite, SOM, river sediments Agricultural soil Limestone, alumina, Silica HA, soil Soil Soil Limestone Limestone Limestone Montmorillonite SOM Kaolinite, soil Biosolids-amended soils   | (D'Alessio et al., 2019; Srinivasan et al., 2013) (Chessa et al., 2016; Goulas et al., 2018; Liu et al., 2017a; Liu et al., 2019; Pereira Leal et al., 2013; Vasudevan et al., 2009; Zhang et al., 2019c; Zhao et al. 2018) (Sukul et al., 2008) (Goyne et al., 2005; Wu et al., 2014) (Gardner and Gunsch, 2017) (Pereira Leal et al., 2013) (Jones et al., 2005) (Tetzner et al., 2016) (Tetzner et al., 2016) (Okaikue-Woodi et al., 2018) (Richter et al., 2009) (Wang and Wang, 2015; Zhao et al., 2011) (Chessa et al., 2016)  |
| Cation bridging                           | Sulfadiazine Ofloxacin Enrofloxacin Enrofloxacin Danofloxacin Oxytetracycline Sulfadimethoxine Sulfacetamide Tetracycline Sulfathiazole Tetracycline Ciprofloxacin Tetracycline Sulfonamide   | Clay-humic complexes Soil Soil, biosolids-amended soil, kaolinite, montmorillonite, tourmaline, biotite, palygorskite, SOM, river sediments Agricultural soil Limestone, alumina, Silica HA, soil Soil Soil Limestone Limestone Limestone Montmorillonite SOM Kaolinite, soil Biosolids-amended soils Organo-montmorillonites, ferrihydrite, soil, goethite, HA Soil  | (D'Alessio et al., 2019; Srinivasan et al., 2013) (Chessa et al., 2016; Goulas et al., 2018; Liu et al., 2017a; Liu et al., 2019; Pereira Leal et al., 2013; Vasudevan et al., 2009; Zhang et al., 2019c; Zhao et al. 2018) (Sukul et al., 2008) (Goyne et al., 2005; Wu et al., 2014) (Gardner and Gunsch, 2017) (Pereira Leal et al., 2013) (Jones et al., 2005) (Tetzner et al., 2016) (Tetzner et al., 2016) (Okaikue-Woodi et al., 2018) (Richter et al., 2009) (Wang and Wang, 2015; Zhao et al., 2011) (Chessa et al., 2016) (Christl et al., 2016; Liu et al., 2012; Wang and Wang, 2015; Zhou et al., 2016) (Zhang et al., 2019b)   |
|   | Sulfamethoxazole<br>Ciprofloxacin  Sulfadiazine Ofloxacin Enrofloxacin Danofloxacin Oxytetracycline Sulfadimethoxine Sulfacetamide Tetracycline Sulfathiazole Tetracycline Ciprofloxacin Tetracycline Sulfamethoxazole  | Clay-humic complexes Soil Soil, biosolids-amended soil, kaolinite, montmorillonite, tourmaline, biotite, palygorskite, SOM, river sediments Agricultural soil Limestone, alumina, Silica HA, soil Soil Soil Limestone Limestone Limestone Montmorillonite SOM Kaolinite, soil Biosolids-amended soils Organo-montmorillonites, ferrihydrite, soil, goethite, HA Soil Agricultural soil, HA, soil  | (D'Alessio et al., 2019; Srinivasan et al., 2013) (Chessa et al., 2016; Goulas et al., 2018; Liu et al., 2017a; Liu et al., 2019; Pereira Leal et al., 2013; Vasudevan et al., 2009; Zhang et al., 2019c; Zhao et al. 2018) (Sukul et al., 2008) (Goyne et al., 2005; Wu et al., 2014) (Gardner and Gunsch, 2017) (Pereira Leal et al., 2013) (Jones et al., 2005) (Tetzner et al., 2016) (Tetzner et al., 2016) (Okaikue-Woodi et al., 2018) (Richter et al., 2009) (Wang and Wang, 2015; Zhao et al., 2011) (Chessa et al., 2016) (Christl et al., 2016; Liu et al., 2012; Wang and Wang, 2015; Zhou et al., 2016) (Zhang et al., 2019b) (Qin et al., 2016; Zhang et al., 2019b; Zhao et al., 2018)  |
| Electrostatic                             | Sulfadiazine Ofloxacin Enrofloxacin Enrofloxacin Danofloxacin Oxytetracycline Sulfadimethoxine Sulfacetamide Tetracycline Sulfathiazole Tetracycline Ciprofloxacin Tetracycline Sulfonamide   | Clay-humic complexes Soil Soil, biosolids-amended soil, kaolinite, montmorillonite, tourmaline, biotite, palygorskite, SOM, river sediments Agricultural soil Limestone, alumina, Silica HA, soil Soil Soil Limestone Limestone Limestone Montmorillonite SOM Kaolinite, soil Biosolids-amended soils Organo-montmorillonites, ferrihydrite, soil, goethite, HA Soil Agricultural soil, HA, soil Palygorskite-montmorillonite, biotite, tourmaline,   | (D'Alessio et al., 2019; Srinivasan et al., 2013) (Chessa et al., 2016; Goulas et al., 2018; Liu et al., 2017a; Liu et al., 2019; Pereira Leal et al., 2013; Vasudevan et al., 2009; Zhang et al., 2019c; Zhao et al. 2018) (Sukul et al., 2008) (Goyne et al., 2005; Wu et al., 2014) (Gardner and Gunsch, 2017) (Pereira Leal et al., 2013) (Jones et al., 2005) (Tetzner et al., 2016) (Tetzner et al., 2016) (Okaikue-Woodi et al., 2018) (Richter et al., 2009) (Wang and Wang, 2015; Zhao et al., 2011) (Chessa et al., 2016) (Christl et al., 2016; Liu et al., 2012; Wang and Wang, 2015; Zhou et al., 2016) (Zhang et al., 2019b)   |
|   | Sulfamethoxazole<br>Ciprofloxacin  Sulfadiazine Ofloxacin Enrofloxacin Danofloxacin Oxytetracycline Sulfadimethoxine Sulfacetamide Tetracycline Sulfathiazole Tetracycline Ciprofloxacin  Tetracycline Sulfonamide Sulfonamide Sulfonamide Sulfamethoxazole Ciprofloxacin | Clay-humic complexes Soil Soil, biosolids-amended soil, kaolinite, montmorillonite, tourmaline, biotite, palygorskite, SOM, river sediments Agricultural soil Limestone, alumina, Silica HA, soil Soil Soil Limestone Limestone Limestone Montmorillonite SOM Kaolinite, soil Biosolids-amended soils Organo-montmorillonites, ferrihydrite, soil, goethite, HA Soil Agricultural soil, HA, soil Palygorskite-montmorillonite, biotite, tourmaline, halloysite, biotite                             | (D'Alessio et al., 2019; Srinivasan et al., 2013) (Chessa et al., 2016; Goulas et al., 2018; Liu et al., 2017a; Liu et al., 2019; Pereira Leal et al., 2013; Vasudevan et al., 2009; Zhang et al., 2019c; Zhao et al. 2018) (Sukul et al., 2008) (Goyne et al., 2005; Wu et al., 2014) (Gardner and Gunsch, 2017) (Pereira Leal et al., 2013) (Jones et al., 2005) (Tetzner et al., 2016) (Tetzner et al., 2016) (Okaikue-Woodi et al., 2018) (Richter et al., 2009) (Wang and Wang, 2015; Zhao et al., 2011) (Chessa et al., 2016) (Christl et al., 2016; Liu et al., 2012; Wang and Wang, 2015; Zhou et al., 2016) (Zhang et al., 2019b) (Qin et al., 2016; Zhang et al., 2019b; Zhao et al., 2018) (Liu et al., 2019; Quan and Bi, 2017)  |
| Electrostatic                             | Sulfadiazine Ofloxacin Enrofloxacin Enrofloxacin Danofloxacin Oxytetracycline Sulfadiamethoxine Sulfacetamide Tetracycline Sulfathiazole Tetracycline Ciprofloxacin Tetracycline Sulfathazole Ciprofloxacin Sulfonamide Sulfamethoxazole Ciprofloxacin Sulfadiazine       | Clay-humic complexes Soil Soil, biosolids-amended soil, kaolinite, montmorillonite, tourmaline, biotite, palygorskite, SOM, river sediments Agricultural soil Limestone, alumina, Silica HA, soil Soil Soil Limestone Limestone Limestone Limestone Montmorillonite SOM Kaolinite, soil Biosolids-amended soils Organo-montmorillonites, ferrihydrite, soil, goethite, HA Soil Agricultural soil, HA, soil Palygorskite-montmorillonite, biotite, tourmaline, halloysite, biotite Agricultural soil | (D'Alessio et al., 2019; Srinivasan et al., 2013) (Chessa et al., 2016; Goulas et al., 2018; Liu et al., 2017a; Liu et al., 2019; Pereira Leal et al., 2013; Vasudevan et al., 2009; Zhang et al., 2019c; Zhao et al. 2018) (Sukul et al., 2008) (Goyne et al., 2005; Wu et al., 2014) (Gardner and Gunsch, 2017) (Pereira Leal et al., 2013) (Jones et al., 2005) (Tetzner et al., 2016) (Okaikue-Woodi et al., 2018) (Richter et al., 2009) (Wang and Wang, 2015; Zhao et al., 2011) (Chessa et al., 2016) (Christl et al., 2016; Liu et al., 2012; Wang and Wang, 2015; Zhou et al., 2016) (Zhang et al., 2019b) (Qin et al., 2019; Quan and Bi, 2017) (Qin et al., 2016)   |
| Cation bridging  Electrostatic attraction | Sulfamethoxazole<br>Ciprofloxacin  Sulfadiazine Ofloxacin Enrofloxacin Danofloxacin Oxytetracycline Sulfadimethoxine Sulfacetamide Tetracycline Sulfathiazole Tetracycline Ciprofloxacin  Tetracycline Sulfonamide Sulfonamide Sulfonamide Sulfamethoxazole Ciprofloxacin | Clay-humic complexes Soil Soil, biosolids-amended soil, kaolinite, montmorillonite, tourmaline, biotite, palygorskite, SOM, river sediments Agricultural soil Limestone, alumina, Silica HA, soil Soil Soil Limestone Limestone Limestone Montmorillonite SOM Kaolinite, soil Biosolids-amended soils Organo-montmorillonites, ferrihydrite, soil, goethite, HA Soil Agricultural soil, HA, soil Palygorskite-montmorillonite, biotite, tourmaline, halloysite, biotite                             | (D'Alessio et al., 2019; Srinivasan et al., 2013) (Chessa et al., 2016; Goulas et al., 2018; Liu et al., 2017a; Liu et al., 2019; Pereira Leal et al., 2013; Vasudevan et al., 2009; Zhang et al., 2019c; Zhao et al., 2018) (Sukul et al., 2008) (Goyne et al., 2005; Wu et al., 2014) (Gardner and Gunsch, 2017) (Pereira Leal et al., 2013) (Jones et al., 2005) (Tetzner et al., 2016) (Tetzner et al., 2016) (Okaikue-Woodi et al., 2018) (Richter et al., 2009) (Wang and Wang, 2015; Zhao et al., 2011) (Chessa et al., 2016) (Christl et al., 2016; Liu et al., 2012; Wang and Wang, 2015; Zhou et al., 2016) (Zhang et al., 2019b) (Qin et al., 2016; Zhang et al., 2019b; Zhao et al., 2018) (Liu et al., 2019; Quan and Bi, 2017) |

Table 5 (continued)

| Sorption<br>mechanism | Sorbate   | Sorbent  | Reference  |
|-----------------------|---|--|--|
|                       | Sulfadimethoxine  | Soil   | (Zhang et al., 2019b)  |
|                       | Cephapirin  | Quartz, feldspar   | (Peterson et al., 2009)  |
|                       | Gibberellic   | Ferrihydrite   | (Pan and Chu, 2016)  |
|                       | Spiramycin  | Palygorskite   | (Ma et al., 2019)  |
|                       | Clorithromycin  | НА   | (Christl et al., 2016)   |
|                       | Ofloxacin   | Graphite   | (Hoque et al., 2017)   |
|                       | Tylosin   | Soil   | (Wegst-Uhrich et al., 2014)  |
|                       | Ciprofloxacin   | River sediments  | (Liu et al., 2017a)  |
|                       |   | Aluminum, iron oxides, layered manganese, dioxide  | (Chao et al., 2015; Figueroa et al., 2004; Jiang et al., 2015; Teixido et al., 2012  |
|                       | Tetracycline  | birnessite, montmorillonite, kaolinite, soil, ferrihydrite, goethite   | Wang and Wang, 2015; Zhang et al., 2015; Zhou et al., 2016)  |
|                       | Norfloxacin   | Soil   | (Peruchi et al., 2015)   |
|                       | Sulfamethazine  | HA   | (Guo et al., 2015)   |
|                       | Sulfame tho xazole  | Soil   | (Berhane et al., 2016; Morel et al., 2014)   |
|                       | Sulfapyridine   | Soil   | (Berhane et al., 2016)   |
|                       | Ciprofloxacin   | Soil, goethite, tourmaline, biotite, SOM   | (Goulas et al., 2018; Gu et al., 2015; Liu et al., 2019; Vasudevan et al., 2009)   |
|                       | Sulfadiazine  | Soil   | (Sukul et al., 2008)   |
|                       | Enrofloxacin  | HAs, soil  | (Gardner and Gunsch, 2017)   |
|                       | Tylosin   | Goethite, HA   | (Guo et al., 2015; Guo et al., 2014)   |
|                       | Gibberellic   | Ferrihydrite   | (Pan and Chu, 2016)  |
|                       | Nalidixic   | Goethite   | (Wang et al., 2017)  |
|                       | Ofloxacin   | Kaolinite, Smectite  | (Gouza et al., 2017; Yan et al., 2012)   |
|                       | Enrofloxacin  | Smectite clay  | (Yan et al., 2012)   |
|                       | Chlortetracycline   | Goethite   | (Qin et al., 2018)   |
| •                     | Nalidixic acid  | Montmorillonite  | (Wu et al., 2013b)   |
|                       | Acetaminophen   | River sediments  | (Yamamoto et al., 2009)  |
|                       | Atenolol  | River sediments  | (Yamamoto et al., 2009)  |
|                       | Carbamazepine   | River sediments  | (Yamamoto et al., 2009)  |
|                       | Ibuprofen   | River sediments  | (Yamamoto et al., 2009)  |
|                       | Ifenprodil  | River sediments  | (Yamamoto et al., 2009)  |
|                       | Indomethacin  | River sediments  | (Yamamoto et al., 2009)  |
|                       | Mefenamic acid  | River sediments  | (Yamamoto et al., 2009)  |
|                       | Propranolol   | River sediments  | (Yamamoto et al., 2009)  |
|                       | β-blocker   | Natural sediment   | (Martinez-Hernandez et al., 2014)  |
|                       | Atenolol  | Natural sediment   | (Martinez-Hernandez et al., 2014)  |
|                       | Stimulant   | Natural sediment   | (Martinez-Hernandez et al., 2014)  |
|                       | Caffeine<br>Norfloxacin   | Natural sediment<br>Wetland soil   | (Martinez-Hernandez et al., 2014)  |
|                       | Ciprofloxacin   | Wetland soil   | (Conkle et al., 2010)<br>(Conkle et al., 2010)   |
|                       | Ofloxacin   | Wetland soil   | (Conkle et al., 2010)  |
|                       | Sulfamethazine  | Goethite   | (Biel-Maeso et al., 2019)  |
|                       | Sulfamethoxazole  | Agricultural soil  | (Qin et al., 2016)   |
|                       | Sulfadiazine  | Agricultural soil  | (Qin et al., 2016)   |
|                       | Sulfadimidine   | SOM  | (Tri et al., 2018)   |
|                       | Oxytetracycline   | Sedimentary soil   | (Xu et al., 2016)  |
|                       | Levofloxacin  | Goethite   | (Wang et al., 2018; Wang et al., 2019)   |
|                       | Sulfamethoxazole  |  | (Ji et al., 2009)  |
|                       | Sulfapyridine   | Graphite   | (Ji et al., 2009)  |
|                       | Tetracycline  | Graphite   | (Ji et al., 2009)  |
|                       | Sulfamethazine  | Goethite   | (Tri et al., 2018; Xu et al., 2017b)   |
|                       | Tetracycline  | Montmorillonite  | (Okaikue-Woodi et al., 2018)   |
|                       | Norfloxacin   | Wetland soil   | (Conkle et al., 2010; Peng et al., 2014)   |
|                       | Sulfamethoxazole  |  | (dos Santos et al., 2017; Qin et al., 2016; Zhang et al., 2011; Zhao et al., 2018)   |
|                       | Ciprofloxacin   | Montmorillonite, kaolinite, goethite, SOM, HA,   | (Conkle et al., 2010; dos Santos et al., 2017; Goulas et al., 2018; Gu et al., 2018  |
|                       |   | wetland soil   | Quan and Bi, 2017; Zhang et al., 2019c)  |
|                       | Sulfadiazine  | SOM, agricultural soil, kaolinite, montmorillonite   | (Qin et al., 2016; Sukul et al., 2008; Zhang et al., 2019c)  |
|                       | Ofloxacin   | Soil, HA, wetland soil, goethite   | (Cheng et al., 2016; Conkle et al., 2010; Hoque et al., 2017; Wu et al., 2014)   |
|                       | Carbamazepine   | Palygorskite-montmorillonite, soil   | (Narimannejad et al., 2019; Quan and Bi, 2017)   |
|                       | Tylosin   | Soil, goethite, diatomaceous earth   | (Guo et al., 2014; Sassman et al., 2007; Sun et al., 2019; Wegst-Uhrich et al., 2014)  |
|                       |   | ** **  | (Li et al., 2019a)   |
|                       | Chloramphenicol   | Vermiculite  | 141 C. Min 401301  |
|                       | Chloramphenicol   | Vermiculite Ferrihydrite   | ,  |
| Hydrogen-bonding      | Gibberellic   | Ferrihydrite   | (Pan and Chu, 2016)  |
| Hydrogen-bonding      | Gibberellic<br>Trimethoprim   | Ferrihydrite<br>Soil   | (Pan and Chu, 2016)<br>(Narimannejad et al., 2019)   |
| Hydrogen-bonding      | Gibberellic<br>Trimethoprim<br>Nalidixic  | Ferrihydrite<br>Soil<br>Goethite   | (Pan and Chu, 2016)<br>(Narimannejad et al., 2019)<br>(Li et al., 2017a)   |
| Hydrogen-bonding      | Gibberellic<br>Trimethoprim<br>Nalidixic<br>Tetracycline  | Ferrihydrite<br>Soil<br>Goethite<br>HA   | (Pan and Chu, 2016)<br>(Narimannejad et al., 2019)<br>(Li et al., 2017a)<br>(Christl et al., 2016)   |
| Hydrogen-bonding      | Gibberellic<br>Trimethoprim<br>Nalidixic<br>Tetracycline<br>Sulfamethoxazole  | Ferrihydrite<br>Soil<br>Goethite<br>HA<br>HA   | (Pan and Chu, 2016)<br>(Narimannejad et al., 2019)<br>(Li et al., 2017a)<br>(Christl et al., 2016)<br>(dos Santos et al., 2017)  |
| Hydrogen-bonding      | Gibberellic<br>Trimethoprim<br>Nalidixic<br>Tetracycline<br>Sulfamethoxazole<br>Ciprofloxacin   | Ferrihydrite<br>Soil<br>Goethite<br>HA<br>HA<br>HA   | (Pan and Chu, 2016) (Narimannejad et al., 2019) (Li et al., 2017a) (Christl et al., 2016) (dos Santos et al., 2017) (dos Santos et al., 2017)  |
| Hydrogen-bonding      | Gibberellic<br>Trimethoprim<br>Nalidixic<br>Tetracycline<br>Sulfamethoxazole<br>Ciprofloxacin<br>Clorithromycin   | Ferrihydrite<br>Soil<br>Goethite<br>HA<br>HA<br>HA   | (Pan and Chu, 2016) (Narimannejad et al., 2019) (Li et al., 2017a) (Christl et al., 2016) (dos Santos et al., 2017) (dos Santos et al., 2017) (Christl et al., 2016)   |
| Hydrogen-bonding      | Gibberellic<br>Trimethoprim<br>Nalidixic<br>Tetracycline<br>Sulfamethoxazole<br>Ciprofloxacin<br>Clorithromycin<br>Pyraoxystrobin                                   | Ferrihydrite Soil Goethite HA HA HA Soil   | (Pan and Chu, 2016) (Narimannejad et al., 2019) (Li et al., 2017a) (Christl et al., 2016) (dos Santos et al., 2017) (dos Santos et al., 2017) (Christl et al., 2016) (Yuan et al., 2019)   |
| Hydrogen-bonding      | Gibberellic<br>Trimethoprim<br>Nalidixic<br>Tetracycline<br>Sulfamethoxazole<br>Ciprofloxacin<br>Clorithromycin<br>Pyraoxystrobin<br>Chloramphenicol                | Ferrihydrite Soil Goethite HA HA HA Soil Vermiculite   | (Pan and Chu, 2016) (Narimannejad et al., 2019) (Li et al., 2017a) (Christl et al., 2016) (dos Santos et al., 2017) (dos Santos et al., 2017) (Christl et al., 2016) (Yuan et al., 2019) (Li et al., 2019a)                                      |
| Hydrogen-bonding      | Gibberellic<br>Trimethoprim<br>Nalidixic<br>Tetracycline<br>Sulfamethoxazole<br>Ciprofloxacin<br>Clorithromycin<br>Pyraoxystrobin<br>Chloramphenicol<br>Bisphenol A | Ferrihydrite Soil Goethite HA HA HA UN HA HA Soil Vermiculite Palygorskite-montmorillonite                       | (Pan and Chu, 2016) (Narimannejad et al., 2019) (Li et al., 2017a) (Christl et al., 2016) (dos Santos et al., 2017) (dos Santos et al., 2017) (Christl et al., 2016) (Yuan et al., 2019) (Li et al., 2019a) (Pan et al., 2009; Wu et al., 2013a) |
| Hydrogen-bonding      | Gibberellic<br>Trimethoprim<br>Nalidixic<br>Tetracycline<br>Sulfamethoxazole<br>Ciprofloxacin<br>Clorithromycin<br>Pyraoxystrobin<br>Chloramphenicol                | Ferrihydrite Soil Goethite HA HA HA UNITIAN SOII Vermiculite Palygorskite-montmorillonite Goethite, HA, goethite | (Pan and Chu, 2016) (Narimannejad et al., 2019) (Li et al., 2017a) (Christl et al., 2016) (dos Santos et al., 2017) (dos Santos et al., 2017) (Christl et al., 2016) (Yuan et al., 2019) (Li et al., 2019a)                                      |

Table 5 (continued)

| Sorption<br>mechanism | Sorbate                  | Sorbent   | Reference   |
|-----------------------|--------------------------|---|---|
|                       | Acetaminophen            | Sediment soil, aquifer sand                               | (Lorphensri et al., 2007)   |
|                       | Atenolol                 | Sediment, soil  | (Yamamoto et al., 2009)   |
|                       | Ifenprodil               | Sediment, soil  | (Yamamoto et al., 2009)   |
|                       | Indomethacin             | Sediment, soil  | (Yamamoto et al., 2009)   |
|                       | Mefenamic acid           | Sediment, soil  | (Yamamoto et al., 2009)   |
|                       | Propranolol              | Sediment, soil  | (Yamamoto et al., 2009)   |
|                       | Carbamazepine            | Sediment, soil  | (Yamamoto et al., 2009)   |
|                       | Ibuprofen                | Sediment, soil  | (Yamamoto et al., 2009)   |
|                       | Tylosin                  | Montmorillonite, kaolinite, soils, HA, diatomaceous earth | (Guo et al., 2015; Sassman et al., 2007; Sun et al., 2019; Zhang et al., 2013b) |
|                       | 17α-ethynyl<br>Estradiol | Aquifer sand  | (Lorphensri et al., 2007)   |
|                       | Nalidixic acid           | Montmorillonite, kaolinite                                | (Wu et al., 2013b)  |
|                       | Oxytetracycline          | Montmorillonite   | (Chang et al., 2009b)   |
|                       | Levofloxacin             | Goethite  | (Wang et al., 2019)   |
|                       | Sulfamethoxazole         | Soil  | (Berhane et al., 2016; Pereira Leal et al., 2013)                               |
|                       | Sulfapyridine            | Soil  | (Berhane et al., 2016)  |
|                       | Ciprofloxacin            | SOM   | (Goulas et al., 2018)   |
|                       | Sulfathiazole            | Soil  | (Pereira Leal et al., 2013)   |
|                       | Enrofloxacin             | Soil  | (Pereira Leal et al., 2013)   |
|                       | Sulfadimidine            | Soil  | (Pereira Leal et al., 2013)   |
|                       | Oxytetracycline          | Sedimentary soil  | (Xu et al., 2016)   |
|                       | Norfloxacin              | Wetland soil  | (Conkle et al., 2010)   |
|                       | Ciprofloxacin            | Wetland soil  | (Conkle et al., 2010)   |
|                       | Ofloxacin                | Wetland soil  | (Conkle et al., 2010)   |
|                       | Ciprofloxacin            | Biosolids-amended soil                                    | (Chessa et al., 2016)   |
| van der Waals         | Oxytetracycline          | SOM   | (Feng et al., 2016; Ji et al., 2009)  |
| attractions           | Trimethoprim             | Soil  | (Narimannejad et al., 2019)   |
| attractions           | Nalidixic                | Goethite  | (Li et al., 2017a)  |
|                       | Niflumic                 | Goethite  | (Li et al., 2017a; Wang et al., 2017)   |
| Electrostatic         | Tylosin                  | Goethite  | (Guo et al., 2014)  |
| repulsion             | Nalidixic acid           | Montmorillonite, kaolinite                                | (Wu et al., 2013b)  |
| repuision             | Levofloxacin             | Goethite  | (Wang et al., 2018)   |

increase of its concentration, the adsorbed ofloxacin would spread to some common sorption sites with low sorption energy, such as electrostatic attraction area. Several studies also confirmed the above view that electrostatic attraction was the main mechanism of ofloxacin sorption on HA (Li et al., 2017b; Wang et al., 2017; Wu et al., 2014). For the sorption of neutral pharmaceuticals (i.e., acetaminophen,  $17\alpha$ -ethynyl estradiol) to sand layer containing less organic matter, both hydrophobic and electrostatic sorption mechanisms were expected to be important. However, electrostatic attraction had a greater influence on the sorption than hydrophobic partitioning because of high sorption on positively charged alumina but low sorption to hydrophobic medium (Lorphensri et al., 2007).

Electrostatic interactions have a significant effect on sorption via two mechanisms. Firstly, exospheric complexes may form directly through electrostatic attraction. Secondly, electrostatic interactions may act as a driving force to accelerate the formation of ion pairs for solvent surface hydration separation and/or surface hydration shared ion pairs (Pan and Chu, 2016). The sorption of tetracycline on HA was enhanced in the presence of calcium ions and was stronger under alkaline conditions. In the study of Christl et al. (2016), this increase was attributed to electrostatic interactions between the positively charged tetracycline complex and HA.

# 4.1.3. Electrostatic repulsion

Electrostatic repulsion makes an important contribution to limit the sorption of PPCPs on negatively charged sorbents (Kah et al., 2017). Wu et al. (2013a) studied the sorption of sulfamethoxazole by Dianchi Lake sediment with high organic matter content and confirmed that electrostatic repulsion made a great difference in sulfamethoxazole sorption. The fate and transport of PPCPs in the environment are significantly affected by minerals (Gao and Pedersen, 2010; Zhou et al., 2016). It was reported that the distribution coefficient ( $K_d$ ) of tylosin on different soils was 1.7 to 12 L/kg, and was positively correlated with clay content.

Positively charged tylosin can interact with the clay's surface, which has a negative charge. Guo et al. (2014) suggested that the sorption mechanism of tylosin to goethite might be related to electrostatic repulsion.

## 4.2. Cation bridging

Cation bridging arises from the formation of an inner-sphere complex between an exchangeable cation at the sorbent surface and an anionic functional group on the sorbate (Kah et al., 2017). Cation bridging is an important mechanism for natural sorbents to absorb anions. The sorption of cationic molecules is dominated by the negative charge on solid surfaces, such as clay mineral surfaces or organic matter. The anion molecules are finally bonded to the positively charged surface of the soil component (the edges of layered clay, the free radical component of humus) by cation bridging (Wang and Wang, 2015; Zhao et al., 2011). High concentrations of heavy metals and PPCPs in the soil often coexist due to the application of large amounts of animal manure and other inputs (Jia et al., 2008). The sorption of tetracycline on clay minerals was stronger in the presence of Cu<sup>2+</sup> than in the absence of Cu<sup>2+</sup> (Wang and Wang, 2015; Zhao et al., 2011). The results of batch sorption experiments showed that due to the cationic bridging effect, the presence of bivalent metal cation could increase the sorption of tetracycline on montmorillonite (Zhang et al., 2010). In the presence of Cu<sup>2+</sup> (0.2 mM), the sorption of tetracycline on Anthrosol was greatly enhanced at pH 6.5, due to the cationic bridge of Cu<sup>2+</sup> between tetracycline molecules and soil particles (Jia et al., 2008). Sun et al. (2019) showed that cationic bridging occurred when sulfonamides were adsorbed on limestone. Duan et al. (2018) found that the metal oxides in tourmaline, halloysite and biotite could serve as a cationic bridge between the carboxylate groups of ciprofloxacin and the interlayer cations of three minerals. Previous studies have also indicated that the sorption mechanism of ofloxacin and enrofloxacin on organic soil is cation bridging (Goyne et al., 2005; Sukul et al., 2008; Wu et al., 2014).

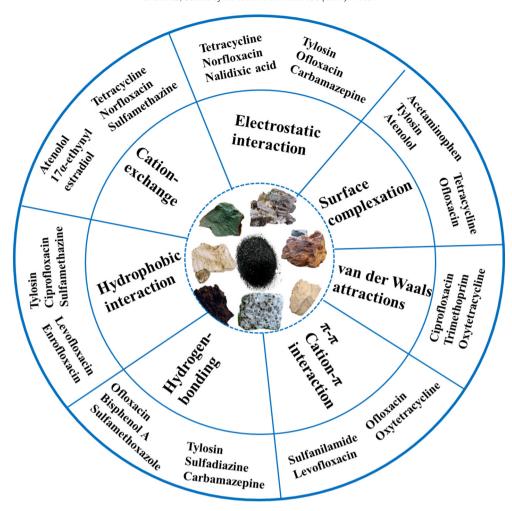


Fig. 2. Sorption mechanisms of PPCPs to soils and components (e.g., SOM, kaolinite, montmorillonite, limestone, goethite). Various PPCPs can be adsorbed to soil and soil components via different mechanisms. The dotted circle inside represents the main components of soil, and each sector includes the sorption mechanism of various PPCPs to soil and soil components.

# 4.3. Surface complexation

Sodium manganese is one of the most common manganese oxides in clay-sized sections ( $<2\,\mu m$ ) with high CEC and large surface area. It was concluded that tetracycline sorption on birnessite occurred mainly through the formation of a horizontal monolayer configuration in the interlayer of birnessite according to X-ray diffraction (XRD) results (Ghadim et al., 2013; Jiang et al., 2015; Li and Wong, 2015). Wu et al. (2019) described the underlying mechanism of interaction between tetracycline and iron oxide minerals (i.e., hydropyrite and goethite) were to form a stable inner spherical complex. Teixido et al. (2012) concluded that tetracycline adsorbed on natural soils by complexing with the sorbent surface. Jiang et al. (2015) also showed that the surface complexation between tetracycline and rectorite was confirmed as the main sorption mechanism through FTIR analysis.

For other PPCPs that can form strong complexes with metallic cations, such as sulfonamides and fluoroquinolones, similar interactions may occur when heavy metals coexist with these PPCPs (Wang et al., 2015). Morel et al. (2014) stated that in the presence of Cu<sup>2+</sup> as a powerful complex agent, the sorption of sulfamethoxazole was substantially strengthened by the formation of a ternary surface complex (Chen et al., 2017). Peruchi et al. (2015) suggested that surface complexation made a significant contribution to the sorption of norfloxacin to soils and there was a tendency to enhance norfloxacin sorption in the soil with higher CEC and clay content.

Some scholars found that tylosin is a weakly basic molecule, whose active functional groups (–OH) can react with those on goethite, and the main sorption interaction of tylosin on goethite may be surface complexation (Guo et al., 2013) Duan et al. (2018) pointed out that a higher sorption capacity of ciprofloxacin to halloysite than to biotite and tourmaline was ascribed to its greater surface area, pore volume and width, conditions that favor more surface complexation interactions with adsorbed ciprofloxacin molecules (Antilen et al., 2016). In soil, ciprofloxacin can be strongly adsorbed, and its sorption coefficient Kd was 210–61,000 L/kg, depending on the nature of the soil (Vasudevan et al., 2009). Gu et al. (2015) showed that ciprofloxacin could directly interact with the surface of goethite through the formation of carboxyl dentate complex, based on spectral data from FTIR and extended X-ray absorption fine structure (EXAFS).

## 4.4. $\pi$ - $\pi$ interaction and cation- $\pi$ bonding

It is considered that  $\pi$ - $\pi$  interactions occur for the neutral forms of aromatic acids (Kah et al., 2017). Tang et al. (2018) studied the complexation of sulfamethazine with Cd<sup>2+</sup> and Pb<sup>2+</sup> on goethite. In their results, cation- $\pi$  interactions may dominate the complex interactions between sulfamethazine and Cd<sup>2+</sup> in acidic environments. Hu et al. (2019) verified through FTIR analysis that the sorption of sulfamethoxazole and sulfadiazine to agricultural soils was dominated by  $\pi$ - $\pi$  interactions. At pH 7, the neutral and anion ratios of sulfamethazine (pK<sub>a</sub>

values of 2.3 and 7.4) were 72% and 28%, respectively. The anion ones were connected to the surface of goethite primarily through  $\pi$ - $\pi$  electron donor-acceptor interaction (Tang et al., 2018). Ahmed et al. (2016) highlighted that one of the main interaction mechanisms of sulfanilamide and SOM was  $\pi$ - $\pi$  interaction. Zhang et al. (2019a) demonstrated the significant role of  $\pi$ - $\pi$  interaction in the sorption of oxytetracycline to SOM. Qin et al. (2016) found that  $\pi$ - $\pi$  interactions between levofloxacin molecules and aromatic rings of organic acids was the main mechanism of increasing sorption of levofloxacin on goethite.

#### 4.5. Hydrogen bonding

Hydrogen bonds in the energy range of 4-17 kJ/mol are formed between negatively charged atoms (e.g., O, N, F) and positively charged H nuclei of functional groups (such as -OH) (Thiele-Bruhn et al., 2004). The strength of a hydrogen bond depends on the discrepancy in proton affinity between the H-donor and H-acceptor atoms. As the distance between the atoms of the donor and the recipient decreases, the bond becomes stronger (Oin et al., 2016; Sukul et al., 2008; Zhang et al., 2019c). Hydrogen bonding is considered to have an important effect on sorption of PPCPs on the soil surface (Kah et al., 2017). The main sorption mechanism of ciprofloxacin and sulfamethoxazole on HA was also proved to be hydrogen bonding of aromatic carboxyl or hydroxyl groups on HA with O atoms in the carbonyl group of ciprofloxacin and amide in sulfamethoxazole (Liu et al., 2017c). Antilen et al. (2016) as well suggested that the sorption of ciprofloxacin on SOM was primarily through hydrogen bonding. H-bonds were suggested to occur for the interactions of ofloxacin and norfloxacin with soil sorbents (Conkle et al., 2010; Wang et al., 2017; Wu et al., 2014; Yu et al., 2019). Xu et al. (2016) used surface plasmon resonance and isothermal titration microcalorimetry analysis to study the dynamics and thermodynamics of the interaction between HA and sulfonamides, and their results showed that hydrogen bonds played an important role in this process. Hu et al. (2019) confirmed through FTIR that the sorption of sulfadiazine and sulfamethoxazole to agricultural soils was dominated by hydrogen bonds. Due to the presence of multiple hydroxyl groups in tylosin molecules, hydrated cations adsorbed on negatively charged soil may make hydrogen bonding a key sorption mechanism for tylosin (Guo et al., 2014; Sassman et al., 2007; Sun et al., 2019; Wegst-Uhrich et al., 2014). Hydrogen-bonding was considered as a sorption mechanism for bisphenol A to palygorskite-montmorillonite, which occurred between the hydroxyl (-OH) or silanol (=Si-OH) groups of palygorskite-montmorillonite and the hydroxyl group of bisphenol A (Berhane et al., 2016).

## 4.6. Hydrophobic interaction

Hydrophobic interaction is a meaningful tractive force for the sorption of sulfonamides and tylosin on soil and soil components (Guo et al., 2014; Pereira Leal et al., 2013; Sassman et al., 2007; Sun et al., 2019; Tetzner et al., 2016; Wang et al., 2019; Wegst-Uhrich et al., 2014). Ji et al. (2009) studied the sorption reaction of sulfamethoxazole and sulfapyridine in sorbents rich in organic matter and found that hydrophobic distribution was the main sorption mechanism of sulfamethoxazole. A speciation-dependent sorption model suggested that hydrophobic interaction accounts for the main part of the sorption of neutral tylosin on clay minerals (Zhang et al., 2013b).

With the increase in the solution pH value, the neutral substance of tylosin dominates. Hydrophobic interactions may dominate the sorption of neutral tylosin on HA, and hydrophobic interactions are also responsible for the sorption of nonionic and less polar organic chemicals on soil. Analogously, within the pH range of 3.0 to 8.0, the neutral form of sulfamethazine was predominant, and the interaction between sulfamethazine and HA mainly depended on hydrophobic distribution (Guo et al., 2015). When the pH value was lower than its  $pK_a$ , nalidixic acid mainly existed as a neutral species, and hydrophobic interaction

resulted in high sorption of nalidixic acid on montmorillonite and kaolinite (Wu et al., 2013b). The hydrophobic interaction between the benzene ring in levofloxacin and the aromatic components of natural organic matter in soil is the main mechanism of the sorption of PPCPs to natural organic matter (Qin et al., 2018). In the research of Chen et al. (2017), the linear sorption isotherm perfectly described the sorption reaction and it was estimated that the main mechanism of sorption of sulfonamide in neutral substances was dominated by hydrophobic interactions. In the presence of background ions (Ca<sup>2+</sup>), hydrophobic interactions decreased when negatively charged substances predominated. (Yamamoto et al., 2009) studied the sorption of eight drugs, including ibuprofen and carbamazepine, on river sediments. The sorption coefficient of selected drugs on river sediments depended on the organic matter content in the sediments. This indicated that hydrophobic interactions between organic matter and drug molecules constituted the main mechanism of sorption to sediments.

#### 4.7. Van der Waals attractions

The energy related to van der Waals forces is comparatively low (in the range of 0.4–4 kJ/mol), but van der Waals forces make an important contribution to interactions between soil sorbents and PPCPs (Kah et al., 2017). It is currently assumed that the sorption of organic pollutants on soil occurs through two disparate steps, i.e., a rapid initial sorption to the surface and then slow diffusion to clay minerals and microporous intercalations (Narimannejad et al., 2019). In the aggregation state of matter, there is a weak attraction between molecules, and the scale of the action energy is generally only a few kilojoules per mole to tens of kilojoules, 1-2 orders of magnitude smaller than the bond energy of chemical bonds, also known as van der Waals gravity or fan force. Sidhu et al. (2019) proposed that ciprofloxacin and azithromycin were attracted to negatively charged silica through the van der Waals force. Similarly, van der Waals forces occurred when oxytetracycline was adsorbed on SOM (Feng et al., 2016). PPCP compounds rarely exist alone in nature, and there may be interactions between various compounds that affect their binding to mineral surfaces. Some scholars studied the binding and sorption mechanism of two typical anti-inflammatory drugs (nalidixic and niflumic) on the surface of goethite (Xu et al., 2017a; Xu et al., 2017b). The formation of a nalidixic-niflumic dimer driven by van der Waals interactions enhanced their sorption. Sorption of nonionic molecules was controlled by the distribution of weak van der Waals forces to SOM (Klement et al., 2018).

# 5. Conclusion

This review assembles data on the sorption of PPCPs on soil and soil components collected over the past decade. The sorption process is generally influenced by pH value, IS, temperature and HA. As an important component of soil, humus not only can be used as sorbent, but also can affect the sorption process by changing the characteristics of other sorbents (e.g., surface charge and sorption site). The influence of temperature on the sorption process is mainly reflected in endothermic, exothermic and molecular diffusion rates. Ion strength and pH have significant effects on sorption as well. Ion strength can promote the sorption of PPCPs through cation bridges, and also influences the sorption mechanism and surface charge of sorbents, showing adverse effects on sorption through competitive sorption. According to the  $pK_a$  of PPCPs and solution pH values, PPCPs can exist as nonions, anion, cation and amphoteric ions. The sorption of nonionic compounds to soil components is controlled by their hydrophobicity, mainly through hydrophobic interactions, electron donor-acceptor interactions, and weak van der Waals forces. Ionic compounds can be adsorbed by electrostatic interaction, cation exchange, cationic bridging, complexation, and hydrogen bonding. These findings have significant implications for understanding the fate, migration, and bioavailability of PPCPs in the soil environment.

Overall, previous studies have greatly improved our knowledge of the interaction between PPCPs and soils. On the basis of these findings, the future research is warranted to investigate the sorption behaviors and mechanisms of newly rising PPCPs, and more environmentfriendly sorbents with high removal efficiency are needed to remediate PPCP-polluted soils.

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#### **Declaration of competing interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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