



Review

Veterinary pharmaceutical residues from natural water to tap water: Sales, occurrence and fate



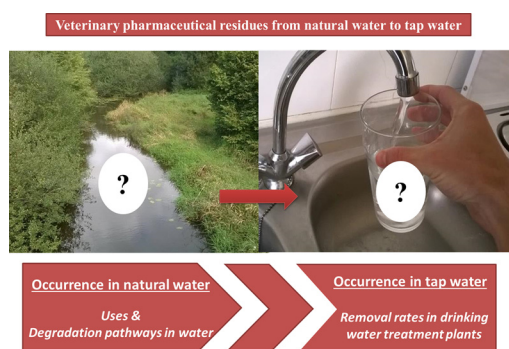
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GRAPHICAL ABSTRACT



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ABSTRACT

Veterinary pharmaceuticals (VPs) increasingly used in animal husbandry have led to their presence in aquatic environments – surface water (SW) or groundwater (GW) – and even in tap water. This review focuses on studies from 2007 to 2017. Sixty-eight different veterinary pharmaceutical residues (VPRs) have been quantified worldwide in natural waters at concentrations ranging from nanograms per liter (ng L^{-1}) to several micrograms per liter ($\mu\text{g L}^{-1}$). An extensive up-to-date on sales and tonnages of VPs worldwide has been performed. Tetracyclines (TCs) antibiotics are the most sold veterinary pharmaceuticals worldwide. An overview of VPRs degradation pathways in natural waters is provided. VPRs can be degraded or transformed by biodegradation, hydrolysis or photolysis. Photo-degradation appears to be the major degradation pathway in SW. This review then reports occurrences of VPRs found in tap water, and presents data on VPRs removal in drinking water treatment plants (DWTPs) at each step of the process. VPRs have been quantified in tap water at ng L^{-1} concentration levels in four studies of the eleven studies dealing with VPRs occurrence in tap water. Overall removals of VPRs in DWTPs generally exceed 90% and advanced treatment processes (oxidation processes, adsorption on activated carbon, membrane filtration) greatly contribute to these removals. However, studies performed on full-scale DWTPs are scarce. A large majority of fate studies in DWTPs have been conducted under laboratory at environmentally irrelevant conditions (high concentration of VPRs (mg L^{-1}), use of deionized water instead of natural water, high concentration of oxidant, high contact time etc.). Also, studies on VPRs occurrence and fate in tap water focus on antibiotics. There is a scientific gap on the occurrence and fate of antiparasitic drugs in tap waters.

Abbreviations: VPs, veterinary pharmaceuticals; VPRs, veterinary pharmaceutical residues

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1. Introduction

As the human population is growing, animal production is also rapidly expanding to feed this population [1]. This leads to intensive agricultural practices, with confined animal feeding operations (CAFOs) [2,3]. In CAFOs, it is a common practice to use veterinary pharmaceuticals (VPs) not only for curative ends, but also prophylactic purposes [1,4,5]. VPs have also been widely used for growth promotion [4], even if this practice has begun to be restricted or even banned, for example in the European Union (EU) [6,7]. Nowadays, more than 2000 veterinary pharmaceutical products are available on the market worldwide [8]. VPRs can be released into the environment either directly with urine and feces of animals in pastures or during aquaculture activities, or indirectly during the spreading of contaminated manure and slurry [7,9–12]. Because of their continuous use, veterinary pharmaceutical residues (VPRs) represent a diffuse and pseudo-persistent pollution in the environment. Among other emerging contaminants, VPRs have been quantified worldwide in aquatic environment, with progresses in analytical methods [10,13,14]. As tap water is produced worldwide from natural waters (SW or GW), it is important to review the occurrence of VPRs in natural waters and the factors affecting this occurrence. Indeed VPRs occurring in natural waters may reach DWTPs which produce tap water. A public health concern is that if these compounds are not fully removed and/or transformed in DWTPs, the human population could be chronically exposed to these compounds. This continuous exposure may represent a risk as VPs are designed to exert biological effects on animals at low doses (as for antibiotic which are active at mg of drug per kg of animal weight) or very low doses (hormones which are active at μg of drug per kg of animal weight) [12]. VPR may also interact together or with other organic contaminants present in drinking water, resulting in addition or synergistic effects [12]. Moreover, chronic exposure of pathogens, commensal or environmental bacteria to veterinary antibiotics can lead to cross-resistance to antibiotics through the development and selection of more harmful bacteria [12,15]. It is a crucial issue as no new class of antibiotics has been discovered since 1987 [8].

However, data on VPRs fate in DWTPs as well as on their occurrence in tap water are scarce and deserve more attention as it is not known if those micropollutants are removed during the applied treatment processes. Lastly, transformation products (TPs) of VPRs should also be considered. Subsequent compounds generated after the transformation process of a certain “parent compound” are considered as TPs. TPs can be generated by natural processes such as metabolization in the treated animals [7,11,16] or via hydrolysis or photolysis in the environment [17–19]. They can also be derived from engineering processes, for example during agricultural waste management practices (i.e. digestion, composting, etc.) [5,20] or in DWTPs during oxidation steps [12,21,22]. These TPs can retain a biological activity [21,23,24] which may pose risks towards aquatic ecosystems or human health.

Ciprofloxacin is the main metabolite of the veterinary antibiotic enrofloxacin, and it is also a pharmaceutical used in human medicine. Both compounds have been proved to exert a negative impact on amphibian larvae growth and development at concentrations greater than or equal to $10 \mu\text{g L}^{-1}$ [25]. Photo-transformation products of the antibiotic tetracycline exhibit a higher toxicity towards luminescent bacteria *V. fischeri* than tetracycline [26] and photoproducts of the hormone altrenogest seem to possess significant androgenic activity according to in vitro tests performed by Wammer et al. [27]. Thus aquatic ecosystems can be negatively impacted by TPs generated by VPRs. When considering TPs produced during tap water production, TPs can gain toxicity during oxidation processes [28,29]. TPs of nitroimidazoles compounds generated during ozonation process were more toxic to *V. fischeri* than the initial nitro-imidazoles [29]. In Leavey-Roback et al. [28], chloramination disinfection of veterinary antibiotics such as tetracyclines or macrolides induces the formation of the carcinogenic disinfection product N-nitrosodimethylamine.

Thus, these TPs may threaten human health.

Many reviews have been published on veterinary pharmaceutical residues in the environment [4,6–9,12,15,16,30–44]. In those reviews, veterinary pharmaceutical residues occurrence and fate in soil have been largely studied, as mobility from soil to water. But only few reviews have been published on occurrence and fate in natural waters (sorption and degradation) and tap water. This review provides an update on research performed from 2007 to 2017 on (1) the occurrence of 65 VPRs (i.e. antibiotics, antiparasitic drugs, anti-inflammatory drugs) in natural waters (surface water and groundwater; 38 studies) related to (2) their uses and their fate. An extensive up-to-date on sales data worldwide, ten years after Sarmah et al., publication [38] is provided, as sales and usages are critical factors impacting environmental occurrence of VPRs [45–47]. Sales trends over the past decade are presented for countries where they are available in the public domain. It is also mentioned when there is no available sales information for countries which are likely to be significant consumers of VPRs. In 2009, Kümmerer [35] reported a lack of information on VPRs degradation pathways, especially on photolysis, with comparison between direct and indirect photolysis. To fill this gap, a complete and up-to-date overview of degradation pathways by chemical family is provided (38 studies). Afterwards, this review focuses on (3) VPRs occurrence in tap water (11 studies) consequently to (4) their behaviour during drinking water treatment plant processes (28 studies).

Few reviews address VPRs behaviour in DWTPs, and deal with pharmaceuticals and personal care products with great emphasis on human pharmaceuticals [23,48] or with antibiotics used both in human and veterinary medicine [32,49]. To our knowledge, this review is the first to gather all available data on VPRs removal rates and fate in DWTPs.

2. Occurrence of VPRs in natural waters

The occurrences of VPRs (since 2007) in surface water and groundwater are presented in Figs. 1 and 2. Fig. 1 focuses on VPRs that have been searched in more than one hundred samples (all studies included). The minimum and maximum quantified concentrations of VPRs in each study are shown on the graphic. Fig. 2 focuses on VPRs searched in less than one hundred samples and shows minimum and maximum concentrations quantified in each study. VPRs that were investigated but not detected have not been reported in Figs. 1 and 2, for example coccidiostat toltrazuril and its metabolites [50], avermectins [51] or some corticosteroids [13,14].

In the 38 studies considered [2,13,14,45–47,51–84], sampling was carried out near husbandry facilities or crops in rural watersheds, once or several times at different seasons of the year.

As reported in Figs. 1 and 2, 64 different VPRs were detected in surface waters, especially rivers [59,61,63,64] but also lakes [55,58], at concentrations ranging from $< 1 \text{ ng L}^{-1}$ [58,68,72] to $255 \mu\text{g L}^{-1}$ (tetracycline [83]; out-of-scale value in Fig. 1). High levels of concentrations depend on the study location and of the veterinarian prescriptions in the corresponding area. For example, the highest concentration level (tetracycline; $255 \mu\text{g L}^{-1}$) was quantified in a sampling site that was assumed to be affected by the discharge of veterinary pharmaceuticals from a swine manure composting facility [83]. Sulfapyridine, sulfamethazine, sulfamethoxy-pyridazine and sulfadiazine were quantified in the Llobregat river in Spain at $12 \mu\text{g L}^{-1}$; 6192 ng L^{-1} ; 3704 ng L^{-1} and 2312 ng L^{-1} , respectively. Similarly, many agricultural areas were located upstream the sampling site [67].

In the 11 studies considered in groundwater, 47 VPRs were quantified with concentrations ranging from $< 1 \text{ ng L}^{-1}$ [76] to $3.461 \mu\text{g L}^{-1}$ for sulfacetamide [79]. Sulfonamides (SAs) constitute the most detected chemical family but also the most studied (28 studies on 38 studies considered) so there may be a bias. Surface water can be a source of contamination for groundwater through artificial or natural exchanges between the two resources [46,59,85].

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