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Fipronil washoff to municipal wastewater from dogs treated with spot-on products

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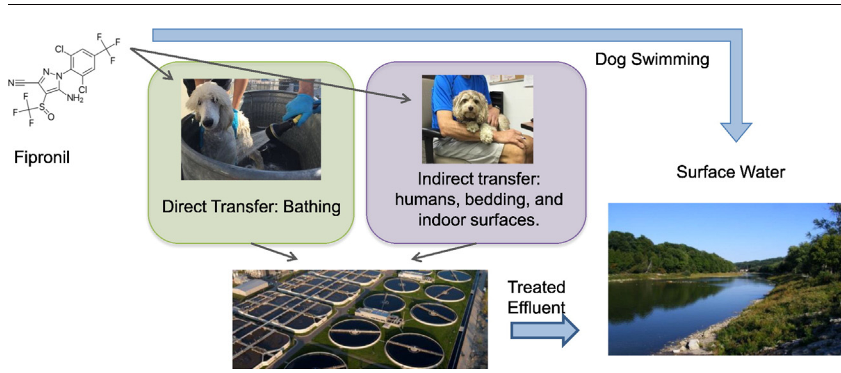
HIGHLIGHTS

- Pathway for pesticide spot-on products to wastewater catchment confirmed.
- Total mass of fiproles measured in rinsate ranged from 3.6–230.6 mg per dog.
- Fipronil spot-on products a source to wastewater influent.
- Fipronil measurable to at least 28 days post application.

GRAPHICAL ABSTRACT



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ABSTRACT

Fipronil and fipronil degradates have been reported in treated wastewater effluent at concentrations that exceed USEPA Aquatic Life Benchmarks, posing a potential risk to the surface waters to which they discharge. Fipronil is a common insecticide found in spot-on flea and tick treatment products that have the potential for down-the-drain transport and direct washoff into surface water. Volunteers currently treating their dogs with a fipronil-containing spot-on product were recruited. Dogs were washed either 2, 7, or 28 days after product application, and rinsate from 34 discrete bathing events were analyzed by LC-MS/MS for fipronil and fipronil degradates (collectively known as fiproles). Total fipronil application dosage ranged from 67.1–410.0 mg per dog following manufacturers' recommendation based on dog body weight. Total mass of fiproles measured in rinsate ranged from 3.6–230.6 mg per dog (0.2– 86.0% of mass applied). Average percentage of fiproles detected in rinsate generally decreased with increasing time from initial application: 21 ± 22, 16 ± 13, and 4 ± 5% respectively for 2, 7, and 28 days post application. Fipronil was the dominant fiprole, >63% of total fiproles for all samples and >92% of total fiproles in 2 and 7 day samples. Results confirm a direct pathway of pesticides to municipal wastewater through the use of spot-on products on dogs and subsequent bathing by either professional groomers or by pet owners in the home. Comparisons of mass loading calculated using California sales data and recent wastewater monitoring results suggest fipronil-containing spot-on products are a potentially important source of fipronil to wastewater treatment systems in California. This study highlights the potential for other active ingredients (i.e., bifenthrin, permethrin, etofenprox, imidacloprid) contained in spot-on and other pet products (i.e., shampoos, sprays) to enter wastewater catchments through bathing activities, posing a potential risk to the aquatic organisms downstream of wastewater discharge.

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1. Introduction

Wastewater treatment plants continuously discharge to rivers, streams, estuaries, and the ocean, carrying contaminants that are not removed during treatment. Arid and semi-arid municipalities struggle to meet demands of urban water use resulting from climate change, population growth and development, leading to increased reliance on wastewater effluent to maintain base flow in urban streams (Luthy et al., 2015). The discharge of treated wastewater effluent to surface water is a major pathway for the introduction of contaminants, including pesticides, to the environment (Luo et al., 2014). Contaminants not removed during treatment, pose a potential risk to aquatic organisms living near or downstream of wastewater outfalls, particularly in water bodies dominated by wastewater effluent. Studies reporting pesticide occurrence in wastewater treatment systems are largely limited to influent and effluent data without information on relative source contribution within a sewershed (Markle et al., 2014; Parry et al., 2015; Sadaria et al., 2016a; 2016b; Supowit et al., 2016; Weston and Lydy, 2010; Weston et al., 2013).

The use of pesticides in outdoor urban areas and subsequent off-site transport to surface water has been documented during storm events (Budd et al., 2015; Ensminger et al., 2013; Thuyet et al., 2012; Weston et al., 2015) and during dry weather conditions as a result of urban irrigation of lawns (Budd et al., 2015; Ensminger et al., 2013; Luo et al., 2013). Resultant surface water pesticide concentrations have frequently exceeded toxicity thresholds resulting in regulatory action by the state of California by both pesticide and water agencies (CDPR, 2012; CVRWQCB, 2017). The majority of U.S. cities rely on separate collection and treatment of stormwater and sanitary discharges; however, some older systems rely on a combined collection system. A 2013 study in Sacramento sampled sub-catchments in the same larger sewershed representing both sole sanitary discharge and combined collection system. Pyrethroid concentrations were comparable in both sub-catchments (Weston et al., 2013), indicating down-the-drain transport of pesticides to sanitary discharge is an important component of total urban mass flux to surface water. Insecticide concentrations (i.e., bifenthrin, permethrin, fipronil, and fipronil sulfone) have been reported at concentrations that exceed USEPA Aquatic Life Benchmarks in treated wastewater effluent (Markle et al., 2014; Sadaria et al., 2016b; USEPA, 2014b). Although the current USEPA Aquatic Life Benchmark for imidacloprid of 1050 ng/L is higher than reported wastewater effluent concentrations (58–306 ng/L) (Sadaria et al., 2016b; USEPA, 2014b), chronic toxicity testing has shown mayfly species are more sensitive to imidacloprid exposures with a reported 28-d EC10 value of approximately 30 ng/L(Roessink et al., 2013; Sadaria et al., 2016b; USEPA, 2014b).

Pesticides used in flea and tick treatments from pet products enter wastewater treatment systems during routine bathing of dogs. Sadaria et al. (2016b) proposed a conceptual model that indicates flea and tick spot-on pet products are the primary source of fipronil and imidacloprid to a wastewater catchment. However, direct measurements of washoff or relative mass flux contribution from sources within a sewershed have not yet been reported. The USEPA is in the process of publishing draft environmental risk assessments for pyrethroids, imidacloprid, and fipronil (December 2016, January 2017, and anticipated summer of 2017 respectively), including the relative contribution from wastewater systems (USEPA, 2017). E-FAST (Exposure and Fate Assessment Screening Tool) is used to predict wastewater effluent concentrations; however, pet spot-on products are not currently included as a source (USEPA, 2014a). At the state level, acting in accordance with the federal Clean Water Act, the Central Valley Regional Water Quality Control Board is in the process of adopting numeric limits for pyrethroids in treated wastewater effluent in response to pyrethroid 303d (impaired water bodies) listings (CVRWQCB, 2017). Developing an understanding of pesticide sources and transport pathways to wastewater treatment catchments is a crucial first step to inform mitigation scenarios and regulatory solutions.

Flea and tick treatments are available with a wide range of pesticide active ingredients (a.i.'s) through several application methods (i.e., spot-on, shampoo, collars, ingestible) for domestic dogs and cats. Dogs are frequently bathed in residential bathtubs, self-serve grooming facilities, or through professional grooming services, where rinsate and dislodged pesticides directly enter a sewer system. Cats are not typically bathed in the same fashion, and thus indirect transfer is a more likely pathway for pesticide residues associated with cat flea and tick treatments to enter the sewershed. The aim of this study is to measure the fraction of fipronil and fipronil degradates, collectively known as fiproles, washed off during routine bathing. However, residues will also be introduced into wastewater treatment catchment through cleaning of indoor surfaces, human showering and washing hands, laundering of materials that have come in contact with pet (i.e., pet bedding, human companion clothes). Studies designed to measure direct human exposure resulting from fipronil spot-on treatments report dislodgeable fiprole residues from a single encounter with a treated pet in the microgram range up to four weeks post application (Cochran et al., 2015; Dyk et al., 2012). Dyk et al. (2012)also quantified pesticide residues on interior surfaces and animal bedding. For the purpose of this study, it was necessary to select a single a.i. and application method to provide a meaningful set of results. Fipronil spot-on products were selected based on parts-per-trillion toxicity of both the parent and degradates and the availability of fipronil containing products (average 8391 kg of dog products per year sold from 2011 to 2015 in California) (CDPR, 2016b).

Fipronil is a phenylpyrazole insecticide registered for uses including structural pest control, bait and gel products, agriculture, and topical flea and tick treatment for pets. In California, fipronil is not registered for agricultural uses. Fiproles are toxic to aquatic invertebrates in the low parts-per-trillion concentration range (Table 1). Fiproles are ubiquitous in San Francisco Bay Area treated wastewater effluent at concentrations that exceed toxicity thresholds posing a risk to aquatic organisms in surface waters receiving discharge (Sadaria et al., 2016b). Detailed studies addressing the removal efficiency of fiproles as a function of specific treatment technology are not available; however, the plants in the above study are all tertiary treatment plants indicating source control, not engineered treatment solutions, may be necessary to reduce effluent concentrations.

The goal of this study is to directly quantify the mass of fiproles washed off volunteer dogs during routine bathing. We compare the measured values to reported wastewater influent monitoring results to investigate the relative contributions from spot-on products to overall sewershed loading. Using available California sales data and commercial shelf survey, we investigate the potential mass transfer of fipronil compared to other a.i,'s. Results will direct future California Department of Pesticide Regulation (CDPR) monitoring efforts.

2. Materials and methods

We solicited volunteer pet owners that were currently using a fipronil containing spot-on product on their dog. Volunteers washed their pet 1–7 days prior to pesticide application, and then applied the product of choice according to the manufacturers' label directions. Pet owners applied the pesticide by squeezing the product from a small applicator onto their pet's neck according to label instructions. Some fraction of the product dose is likely left inside the applicator introducing variability to the total mass applied. Label instructions for the four product brands used by volunteers varied only slightly. All product labels recommend reapplication after 30 days, and indicate products are effective for three months. Frontline Plus™, Petlock Plus™, and Sentry Fiproguard™ labels state the product is waterproof after it has dried and pets can swim and bathe post application. The Pet Armor Plus™ label does not claim to be waterproof. Volunteers reported using one of four fipronil-containing spot-on products. Each manufacturer offers a dose appropriate for pet size (according to body mass), all containing

Table 1
Summary of toxicity and wastewater effluent concentrations reported for pesticides commonly found in pet products.

Compound	Aquatic invertebrates[a] Acute (ng/L)	Chronic (ng/L)	Wastewater effluent (ng/L)
Fipronil	110	11	14–49[b]
Fipronil sulfide	1065	110	1.3–2[b]
Fipronil desulfinyl	100,000	10,300	<0.39–1.2[b]
Fipronil sulfone	360	37	1.1–16.3[b]
Fipronil amide	–	–	<0.2–4.1[b]
Permethrin	10.6	1.4	ND–170[c]
Etofenprox	400	170	NA
S-methoprene	16,500	51,000	NA
Phenothrin	2200	470	NA
Imidacloprid	34,500	1050	83–305[b]

[a] USEPA Aquatic Life Benchmarks (USEPA, 2014b).
[b] (Sadaria et al., 2016b).
[c] (Markle et al., 2014).

8.8–9.1% fipronil. Products come in individually-sized doses with between 67 and 405 mg fipronil per application. Thirty-four dogs were washed in total, with 11, 13, and 10 at 2, 7, and 28 days respectively post application. A complete summary of product types and sizes used on volunteer dogs is found in the supporting information (SI), Table A.1. Several dogs were volunteered for multiple discrete washoff events.

All dogs were weighed (Cardinal Detecto digital scale) and the breed and fur coarseness recorded. Small dogs, roughly <10 kg, were washed in a plastic tub. All larger dogs were washed in a galvanized-metal tub retrofitted with a PVC spout to drain wash-water. Photos are included in SI Figs. A.1–A.3. Between discrete bathing events, the equipment was rinsed with tap water, rinsed with methanol, and finally rinsed with deionized water. Five equipment blanks were collected from the sampling equipment throughout the study.

On the designated day post application, each dog was thoroughly wetted with tap water. Shampoo (WAHL Home Products™ Oatmeal Formula product used throughout study) was then applied to provide lather over the entire animal (volume of shampoo recorded). Following lather, each animal was thoroughly rinsed with tap water. The entire rinsate, including the water added to initially wet the animal, was considered a single sample. The volume of rinsate for small dogs was determined using the mass of the plastic container before and after water collection. After washing large dogs, the rinsate volume was discharged to a plastic basin using a 1-L volumetric beaker to record the volume. The sample volume, soap volume, and dog mass are reported in the SI. First, a 500 mL sub-sample was collected from the entire composite washoff for analysis of fiproles in a glass amber bottle. A 1-L sample was also collected for analysis of total suspended solids (TSS) (Ensminger, 2016). Water quality parameters of rinsate were measured using a YSI Sonde (YSI EXO1).

Chemical analysis of fipronil and degradates was conducted at the California Department of Food and Agriculture's Environmental Safety Lab. A 10-mL aqueous sample is diluted with deionized water to volume of 100 mL before liquid-liquid extraction. Each sample was placed into a 250-mL separatory funnel with 50 mL of methylene chloride and shaken for two minutes. The methylene chloride phase was poured over 70 g of anhydrous sodium sulfate to remove residual water. Extraction steps were repeated two subsequent rounds. The anhydrous sodium sulfate was rinsed with an additional 40 mL of methylene chloride. The resultant extract was evaporated to dryness on a rotary evaporator with a water bath at 30 ± 1 °C and a vacuum maintained at 0.44 bars of mercury. Samples were reconstituted with acetone to a final volume of 1.0 mL. A 5-μL aliquot of extracts was analyzed by liquid chromatography with tandem mass spectrometry (LC-MS/MS) (on an ABSciex QTRAP 5500 Negative Electrospray Ionization (ESI-)).

An untreated dog was washed according to stated protocol to provide a representative shampoo containing matrix for method development and matrix spikes. Triplicate analysis of shampoo containing matrix water spiked at 2, 3, and 5 μg/L with recoveries between 81 and 121%. Method detection limit was developed by analyzing seven matrix spike replicates at 0.5 μg/L. Adopted reporting limits of 1.0 μg/L for fipronil, fipronil sulfide, fipronil sulfone, and fipronil desulfinyl and 1.5 μg/L for fipronil amide and fipronil desulfinyl amide were >10 times the respective method detection limit. Study samples were extracted within three days of sample collection based on acceptable matrix recoveries (80–120%) in spiked samples refrigerated up to three days. Dilutions were made as needed to fit within the calibration range (5–500 μg/L). Further details on instrument and quantification parameters are found in the SI, Tables A.2 and A.3.

3. Calculations

Total mass of fipronil and degradates measured in this study are reported as mass washoff per dog and % washoff per dog (Eqs (1) and (2)). We assume the 500-mL sample is a representative concentration of total rinsate volume. We also assume pet owners applied the entire pesticide dose with negligible residue remaining in the product applicator.

$$\text{mass washoff per dog } (\mu g) = \text{fiprole concentration}\left[\frac{\mu g}{L}\right] \times \text{rinsate}[L] \quad (1)$$

$$\%\text{washoff} = \frac{\text{mass washoff per dog } (\mu g)}{\text{mass applied per dog } (\mu g)} * 100 \quad (2)$$

One objective of the study is to understand the relative contribution of fiproles from pet spot-on treatments to total wastewater fiprole loading to wastewater treatment plants. To compare sources, wastewater monitoring data and spot-on sales data are converted to monthly per capita fiprole loading.

Sadaria et al. (2016b) report service area population and influent fiprole concentrations for seven bay area wastewater treatment plants using 24-h composite samples. Using Eq. (3), a total monthly per capita fiprole load is calculated. We assume fiprole concentrations are representative of a month (30 days). The results are not normalized for pet ownership, but instead we assume an even per capita distribution.

$$\text{total monthly, per capita fiprole load} = \left(\frac{\text{influent}\left[\frac{\mu g}{L}\right] * \text{monthly flow}[L]}{\text{service area population}}\right) \quad (3)$$

An estimate of total monthly spot-on monthly per capita fiprole load is calculated using California statewide sales data from 2011 to 2015 and California population information (Bureau USC, 2016; CDPR, 2016c). The fraction of total fiprole dislodged during bathing is estimated using analytical results from this study and represented by $f_{dislodged}$. Eq. (4) also assumes some fraction (f_{washed}) of treated animals is washed within 28 days of treatment in a location directly plumbed to the sewer.

$$\text{monthly per capita fiproles load from spot-on products} = \frac{\text{sales of spot-on}\left[\frac{\mu g}{month}\right]}{\text{population}} * f_{dislodged} * f_{washed} \quad (4)$$

A ratio of Eqs (4) and (3) represents the relative contribution of spot-on fiproles to total wastewater loading.

4. Results and discussion

Fiproles were detected in 100% of the samples. Results from 34 discrete bathing events are reported as total mass of fiproles (Eq. (1)). Generally, there was a decrease in washable fiprole fraction and a decrease in variability with increasing time post application (Fig. 1). A paired t-test revealed no significant difference between percent washoff of 2 and 7 day samples ($p = 0.246$), but there was a significant difference between both 2 and 28 day samples and 7 and 28 day samples ($p = 0.003$ and 0.0009, respectively). Total fiprole mass recovered ranged from 0.2% to 86% of total mass available (Eqs (1) and (2)). The mass dosage of fipronil in each package ranged from 67 to 405 mg based on the mass of the dog. Total recovered mass of fiproles was between 3.6 and 230.6 mg per dog (using Eq. (1)).

Fipronil and fipronil sulfone were detected in 100% of the samples. Fipronil desulfinyl, fipronil sulfide, and fipronil amide had detection frequencies of 88%, 76%, and 52%, respectively. Desulfinyl fipronil amide was not detected in any of the samples. Fipronil was the dominant form of fiprole and accounted for >63% of total fiproles in all samples and >92% of total fiproles when considering only 2 and 7 days post-application sampling events (Fig. 2). The highest percentage of degradates were found in two discrete 28-day samples collected from the same dog that was reported as having spent all time outdoors. Measured degradates were fipronil sulfone and fipronil desulfinyl, both of which are reported photolysis products (Simon-Delso et al., 2015).There were no trends or relationships observed as a function of dog size. Finally, the measured TSS did not correlate with percent washoff ($r^2 = 0.0131$).

Equipment blank samples contained measurable fipronil in all but one of the samples; however, with mass recovered ranging from 13 to 56 μg fipronil compared to sample recoveries from 113 to 224,900 μg, the potential for carry over is considered insignificant and blank correction calculations were not made. Fipronil amide, fipronil sulfone, and fipronil desulfinyl were measured in some equipment blanks near detection limits. Some fipronil carry-over between samples likely occurred and would be most important for the 28-day samples, which exhibited relatively low overall recoveries.

Fiproles dislodged during routine bathing can enter a wastewater catchment through residential bathtubs, self-serve grooming facilities, and professional grooming facilities. In order to provide some perspective on reported washoff percentage, a comparison between per capita fiprole concentrations based on (1) wastewater monitoring concentrations and an (2) product sales data are provided using Eqs (3) and (4). A recent study by Sadaria et al., (2016b) and others measured fipronil and fipronil degradates entering seven wastewater treatment plants, six serving residential municipalities in the San Francisco Bay Area. Using Eq. (3), the reported total influent fiprole concentrations for six plants are transformed to mass contributed per month per person (with an average of 0.71 ± 0.11 mg fiproles/person/month).

Rather than calculate a single per capita contribution from sales data we present a range of values to characterize the range of possibilities using Eq. (4). California sales data report an average of 8390 kg per year of fipronil sold in the form of spot-on dog treatment from 2011 to 2015 (CDPR, 2016c). The California 2015 State Census reported a population of 39,144,818 (Bureau USC, 2016). For $f_{dislodged}$ we use 0.21, 0.16, and 0.04 to represent average observed wash-off during 2-day, 7-day, and 28-day time points respectively measured during this study. There is no reliable data to inform f_{washed}, or the estimate of for the fraction of fipronil treated dogs washed within 28 days of treatment in locations (i.e., residential bathtubs, self-serve grooming facilities, and professional grooming facilities) that discharge to wastewater catchments (Fig. 3). The authors present this range of values to demonstrate the importance of spot-on pesticide products to overall sewershed loading.

Using this approach, we can see that washing 25% of treated dogs within 7 days of treatment would account for the entire fiprole load in the sewershed. Results suggest spot-on products are an important source of fiproles to wastewater treatment plants. Treated wastewater effluent in the same Northern California study reported fipronil concentrations between 14 and 45 ng/L, which are above the USEPA chronic aquatic benchmark for fipronil (11 ng/L) (Table 1).

Additional mass from both cats and dogs treated with flea and tick treatments can enter wastewater treatment plants from cleaning activities. Fipronil concentrations have been reported on indoor residential dust, and homes with a dog treated with a fipronil-containing spot-on products resulted in 2 to 3 orders of magnitude higher concentration in dust than comparable households that did not have treated pets (Mahler et al., 2009). The transport pathway of organic chemicals bound to household dust to wastewater treatment plants has been confirmed using flame retardant concentrations in household dust and laundry rinsate (Schreder and La Guardia, 2014). Further, human contact with treated pets can lead to down-the-drain transport of fiproles through showering, washing hands, and human excrement.

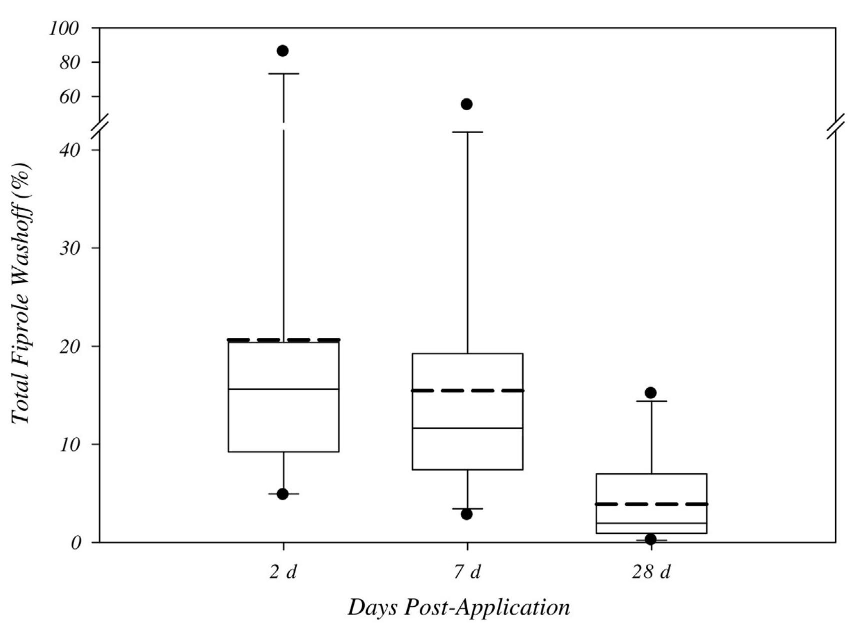

Fig. 1. Percent wash-off of total fiproles as a function of time. Number of discrete samples is 11, 13 and 10. The box encloses the 25th to 75th percentile, whiskers note 5th and 95th percentile, median black solid line dissecting box, blue dashed line the mean, and black circle minimum and maximum.

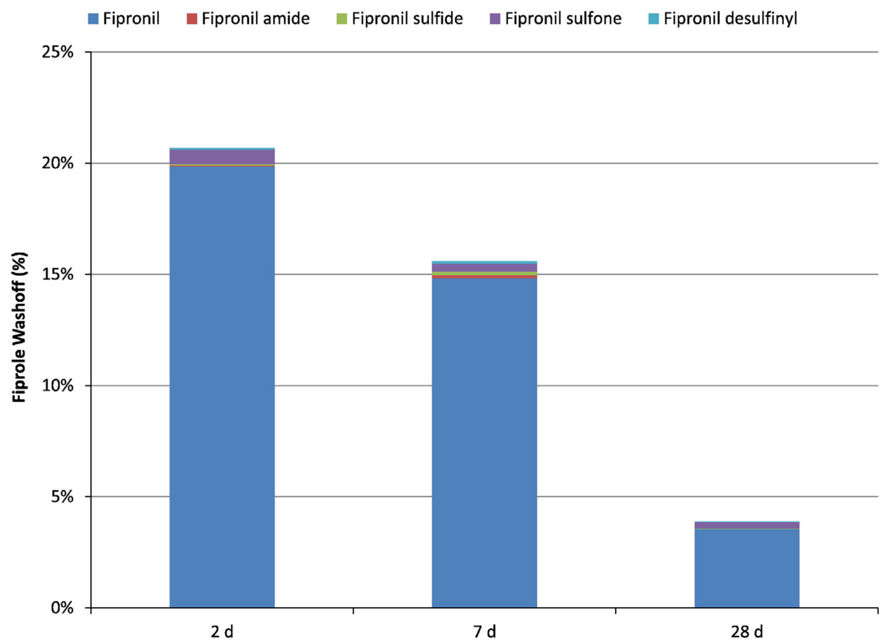

Fig. 2. Average percent fipronil and degradate washoff at two, seven, and twenty-eight days post-application. Values expressed as percent total mass applied.

4.1. Other active ingredients

Direct quantification of washoff potentials for the many spot-on products containing other a.i.'s is beyond the scope of this study. However, California pesticide sales data for 2011–2015 identify fipronil, permethrin, imidacloprid, etofenprox, phenothrin, and s-methoprene as the most common a.i.'s used in spot-on products by mass (Fig. 4) (CDPR, 2016b). A 2014 shelf-survey conducted in the Sacramento region identified 99 pesticide products for pets available to the consumer (34 spot-on products, 14 collars, 28 grooming products, and 23 sprays) (Vander Werf et al., 2015). In addition to the a.i.'s listed above, pet products contain piperonyl butoxide, propoxur, cyphenothrin, esfenvalerate, tetramethrin, novaluron, prallethrin, tetrachlorvinphos, cyhalothrin, and cypermethrin, many of which have not been measured in municipal wastewater. Monitoring data available for fipronil and permethrin in wastewater effluent suggest treatment processes in place do not reduce pesticide concentrations below toxicity thresholds; therefore, these pesticides pose a potential risk to the surface waters to which they discharge (Table 1), particularly in effluent dominated streams in arid regions and estuaries with limited mixing. As noted in the introduction, the current USEPA Aquatic Life Benchmark for imidacloprid does not consider more recent chronic toxicity testing for

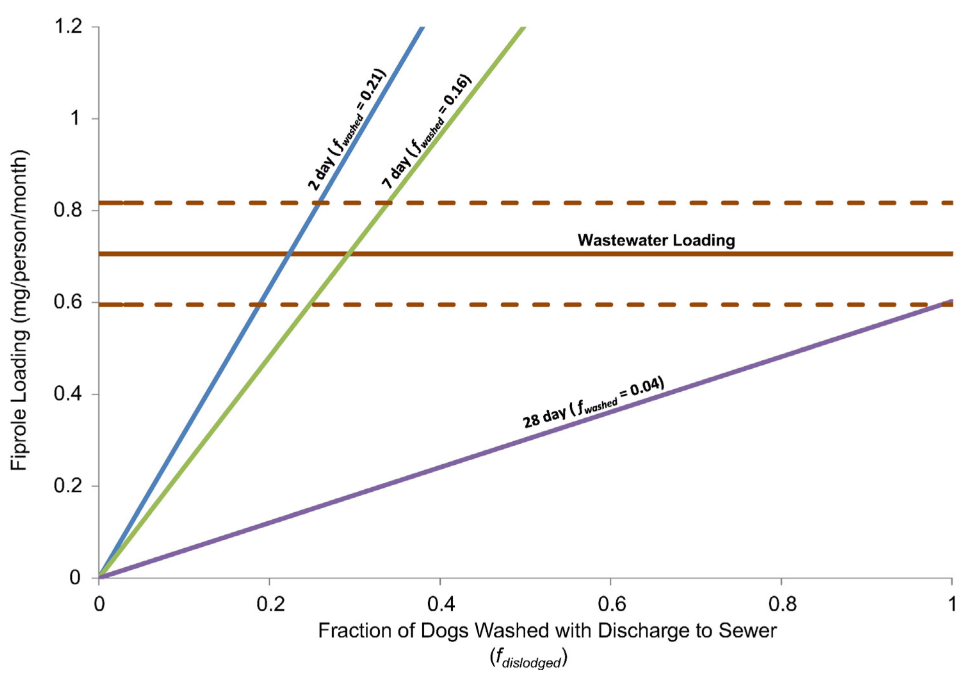

Fig. 3. Comparison between per capita loading using wastewater monitoring data in solid brown (Eq. (3)) and sales data as a function of $f_{dislodged}$ and f_{washed} (Eq. (4)). Dotted wastewater lines represent one standard deviation of Sadaria et al., 2016b dataset (n = 6). The value of $f_{dislodged}$ represents average washoff values measured for 2, 7, and 28 days of 0.21, 0.16, and 0.04, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

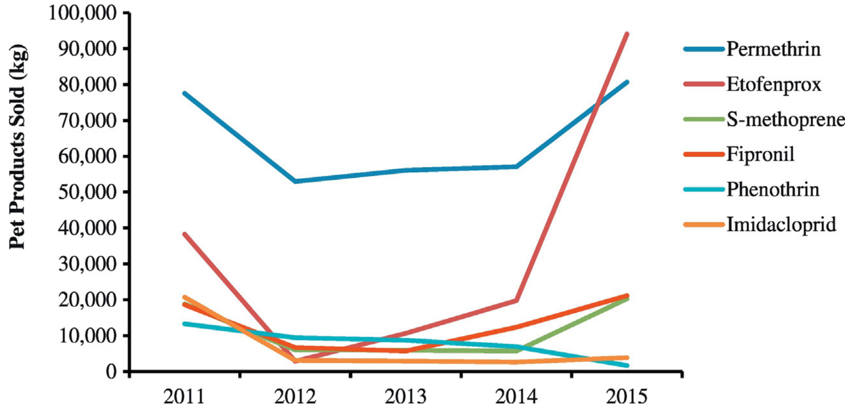

Fig. 4. Kg sold per year of top six pesticides found in pet products from 2011 to 2015. The synergist piperonyl butoxide, and disinfectants dodecyl dimethyl ammonium chloride and alkyl dimethylbenzyl ammonium were excluded from the ranking (CDPR, 2016c).

mayflies. The lack of wastewater effluent data for etofenprox, s-methoprene, and phenothrin represents a data gap that is necessary to fully evaluate the impact of pesticides found in pet products to wastewater effluent.

A.i.'s found in pet products have a wide range of physical and chemical properties that impact initial washoff; however, the pathway has been established. The removal efficiency of specific pesticides during wastewater treatment is still largely unknown. Additional studies are needed to characterize the occurrence and fate of pesticides entering wastewater treatment systems.

5. Conclusion

Fiproles were detected in 100% of the samples up to the 28–day pretreatment interval. Results confirm the down-the-drain transport of pesticides contained in spot-on treatments. Fipronil persisted with little break down to fipronil degradates during the entire 28-day treatment period. At 28 days post application, fiproles can be dislodged and transported down the drain at the magnitude of mg per pet. Measurements of dislodgeable pesticide residues during routine bathing confirm spot-on fipronil treatments contribute a substantial mass fraction of total fipronil loading to the wastewater catchment. The calculated estimates are relatively conservative and do not consider indirect transfer of pesticide residues associated with spot-on residues transported through the cleaning of indoor surfaces, human showering, laundering of materials that have come in contact with pet (i.e., pet bedding, human clothes), and human excrement. Other potential sources of indirect transfer include additional registered uses for fipronil (e.g., indoor crack and crevice, subterranean termite treatments, agriculture (excluding California), urban applications). It is beyond the scope of this study to quantify all potential sources; however, based on our measurements and calculations, spot-on flea and tick treatments have the potential to contribute up to the entire reported wastewater load and thus should be considered as an important source.

Spot-on flea and tick treatments may also be directly transferred to surface water in locations where treated pets swim. The total recovered mass of fiproles was between 3.6 and 230.6 mg per dog. The mass available may pose a risk to small water bodies.

It is beyond the scope of this paper to investigate the human health implications of coming in contact with treated pets; however, it is worth noting that fipronil is the focus of a human health risk assessment initiated by CDPR (CDPR, 2016a). Groomers, children, and adult pet owners may come in contact with fipronil regularly since current product labels do not require personal protective gear during application.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2017.04.219.

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