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Dog swimming and ectoparasiticide water contamination in urban conservation areas: A case study on Hampstead Heath, London

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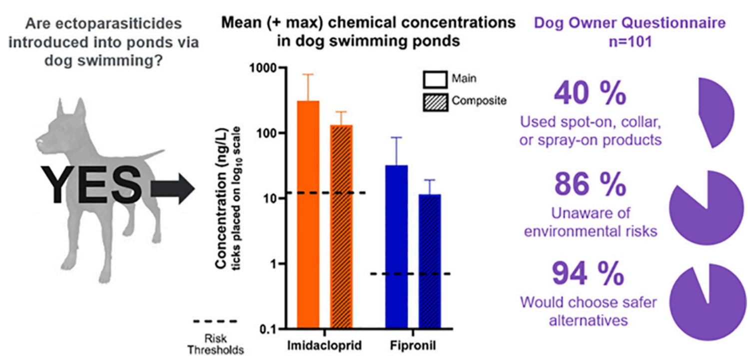
HIGHLIGHTS

- Demonstrated environmental contamination by ectoparasiticides in urban ponds
- Imidacloprid and fipronil in dog ponds exceeded environmental toxicity thresholds
- Concentrations strongly correlated with levels of dog swimming
- Markers of wastewater contamination negligible in source waters
- Survey of owners who swim dogs identified opportunities for risk reduction

GRAPHICAL ABSTRACT



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ABSTRACT

Widespread occurrence of two ectoparasiticide compounds in the aquatic environment, imidacloprid and fipronil, have prompted concerns about their potential environmental impacts. However, very little focus has been placed on water bodies in urban green spaces used for dog swimming. In this study, occurrence of both substances on Hampstead Heath, London, was compared in ponds with ($n = 3$) and without dog swimming activity (n = 3), as well as connecting streams above, between, and below these ponds ($n = 6$). Imidacloprid and fipronil were detected at main swimming points in dog swimming ponds at mean concentrations of 309 ± 104 ng/L and 32 ± 13 ng/L, respectively, indicating a high environmental risk in these samples. Measured concentrations in ponds not accessible for dog swimming were either below the limits of detection or limits of

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quantification for both chemicals. Across all ponds, there was a strong positive correlation between measured dog swimming activities and concentrations of imidacloprid ($R^2 = 0.91$) and fipronil ($R^2 = 0.79$). Some contamination was detected in connecting streams between ponds. A wider chemical analysis for the presence of urban waste water chemical residue signatures indicated minimal contamination, including in source waters. A survey of visitors who allow their dogs to swim in the sampled ponds confirmed frequent use of products containing imidacloprid and fipronil. In total, 86 % of 101 dog owners were unaware of the potential environmental impacts of products, and 94 % indicated that protecting nature would be an important consideration when selecting products. Besides the current practice of limiting dog access to ponds, information collected on product use and dog swimming practices identified additional opportunities to reduce contamination. We suggest that more cooperation between industry, regulators, veterinarians, green space managers, and the public can reduce risks to urban biodiversity while maintaining recreational benefits for dog owners and dogs.

1. Introduction

In the UK, increasing numbers of people are visiting nature-rich green spaces (National Trust, 2020; Office of National Statistics, 2021), and many bring their dogs. Both people and dogs benefit physically, mentally, emotionally, and socially from experiences in green spaces (Zijlema et al., 2019, Koohsari et al., 2022, Gianfredi et al., 2021), but these experiences are not without impact on natural habitats and wildlife (Taylor et al., 2005). Biodiversity impacts to which dogs contribute include disturbance of wildlife (Banks and Bryant, 2007; Beasley et al., 2023), changes in soil chemistry which affect flora (Buchholz et al., 2021, De Frenne et al., 2022), and disturbance to freshwater habitats caused by swimming in ponds and streams (Borgmann, 2011, Bowes et al., 2015).

Many flea and tick treatments applied to dogs include pesticides which could impact insects and other invertebrates in natural habitats (Wells and Collins, 2022). Some of these are applied topically to dogs or impregnated in dog collars and can wash off when dogs enter water bodies (Teerlink et al., 2017; Diepens et al., 2023; Perkins et al., 2024). Two particular compounds, imidacloprid and fipronil, are applied in this manner and have attracted recent attention due to their potential risk to the environment (Preston-Allen et al., 2023). Both are neurotoxic pesticides that are widely used in companion animal ectoparasite products in many parts of the world (Rust et al., 2018; Tyler et al., 2019). Both can have detrimental lethal and sub-lethal effects on non-target aquatic invertebrates and their populations (Pisa et al., 2015, Yamamuro et al., 2019, Merga and Van den Brink, 2021). These chemicals have been shown to be widespread contaminants in English rivers (Perkins et al., 2021). In Europe, their outdoor use for agriculture has not been permitted since 2018 due to their non-target effects, particularly on bees (Preston-Allen et al., 2023; Pesticide Action Network, 2023). However, despite their relatively short half-lives, they are still detected in surface waters. Aside from dietary exposure to humans through contaminated food, any indoor use as a pesticide, or runoff/leaching, veterinary sources of this contamination in the aquatic environment have been indicated (Egli et al., 2023). These can arise, for example, from washing treated animals, contaminated bedding/soft goods, or hands following application, resulting in environmental discharge through wastewater (Perkins et al., 2024). It is also likely that these chemicals are released from treated pets while swimming in rivers and ponds (Teerlink et al., 2017).

While there is experimental evidence that ectoparasiticides are released when treated dogs are immersed in water (Diepens et al., 2023), there is no specific evidence that dogs swimming in nature conservation areas are contaminating waterbodies with ectoparasiticides. For our study, we selected Hampstead Heath, a 275 ha green space in London designated as a Metropolitan Site of Interest for Nature Conservation (City of London Corporation, 2019). The Heath's ancient woodlands, meadows, heathland and spring-fed wetlands are visited by an estimated 10 million people every year (City of London Corporation, 2019). Its many ponds are carefully managed to enhance biodiversity, and some are designated for dog swimming. A distinctive feature of the Heath is the long history of community involvement in its protection as a refuge for both people and nature (Lawrence, 2019). This strong local interest facilitated engagement of the community that regularly swims their dogs on the Heath, who provided information on their use of flea and tick treatments and their attitudes concerning environmental risks.

The aim of this work was to assess the contribution of dog swimming to the detection of selected pet ectoparasiticides in aquatic habitats in protected urban green spaces and to understand dog owners' knowledge and perceptions of the use of flea and tick treatments, their introduction into aquatic ecosystems, and their potential environmental risks. The specific objectives were to: (a) quantitatively analyse pond and connecting stream samples from Hampstead Heath for imidacloprid and fipronil residues; (b) measure a range of other contaminants of emerging concern (CECs) typically observed in urban surface waters, such as pharmaceuticals, illicit drugs and other pesticides; (c) characterise, by questionnaire, the use of flea and tick treatments by dog owners who swim their dogs on Hampstead Heath; and (d) evaluate the awareness of the environmental impact of flea and tick treatments amongst these dog owners. This study investigated site-specific patterns of veterinary flea and tick product use and identified opportunities for risk reduction by local dog owners. This work also provided critical new knowledge and evidence for regulators and those directly responsible for managing green spaces.

2. Materials and methods

2.1. Study location

Hampstead Heath is managed by the City of London Corporation and English Heritage. It contains over 40 distinct waterbodies ranging in size from small ephemeral pools to large reservoirs, and this study concentrated on ponds in two distinct chains as shown in Fig. 1. These ponds are created from rainwater falling on sandy soils and emerging on hillsides at the interface with clay-rich beds (Clemens, 2010). Because this topography lies above the surrounding London landscape, this rain-fed catchment for the Heath's ponds is largely isolated from nearby urban areas and their surface and ground waters (City of London Corporation, 2013). The Heath is a popular area for dog walking and swimming. A survey of Heath visitors in 2017 indicated that 22 % of these came to walk dogs (Groundwork London, 2017). Ponds for dog swimming have operated for decades and have been formally designated since 2010.

Six ponds were chosen for the study; three in which dog swimming is allowed, and three in which dogs are not permitted to swim and where physical access is limited (Fig. 1). Within the three dog swimming ponds (DSPs), dog swimming areas are clearly marked (i.e., main swimming areas): a flat, beach-like area extended into shallow water with a floating barrier at about 15–20 m into the pond that defined an area within which dogs were expected to swim. Dog swimming was discouraged outside these areas (even in the same pond) and in all other ponds, some of which are fenced. However, visitors did occasionally swim their dogs outside of the main swimming area and in restricted ponds. Two of the non-dog swimming ponds (NDSPs) chosen for the study were enclosed by fences and therefore more difficult for dogs to access (A, B).

2.2. Dog activity observations

Dog activity was observed twice, at an interval of two weeks, during the weeks of 12 June 2023 and 26 June 2023. Preliminary, casual observations suggested frequency was greater on weekends than weekdays. Therefore, as an indicator of the frequency of dog swimming in the different ponds per week, dogs entering ponds were recorded on a Wednesday and Saturday of each sampling week. On each day at the three DSPs, observations were made at each pond for five 30-minute sessions between 07:00 and 19:30 BST. Dogs entering the water were classified as 'entry' (dog came into contact with the water) or 'immersion' (dog shoulder was below the water surface). This distinction was made because immersions would specifically expose areas of fur directly treated with spot-on products containing ectoparasiticides. Immersions were a subset of all entries. Additionally, it was noted whether dogs entered the water inside the main swimming area or outside of the main swimming area. Dogs were only counted for their first entry into the pond. Dog swimming at pond D was disrupted during the second sampling week as an algal bloom closed the main dog swimming area; dogs could still enter the pond, but signage discouraged this.

At the three NDSPs, preliminary observations confirmed that dog swimming was extremely infrequent or absent due to fencing and signage. To provide data comparable to DSPs, a point similar in aspect and depth to the designated swimming points in DSPs was selected for each NDSP, and Browning Dark Ops Pro ED (Model BTC-6PXD) motion-sensor cameras (Prometheus Group, Birmingham, AL, USA) were set there to record activity across the pond and at the edge. The cameras were set to trail mode with a 10 second capture delay, and pictures sized up to 16 Mb were captured on SanDisk 32GB Ultra UHS-I SDHC memory cards. (Western Digital Technologies, San Jose, CA, USA). Camera records were taken over the same weekly periods as observations on the DSPs. One of the cameras failed to record during this period, so an additional two weeks were recorded after the data collection period in compensation.

As a measure of dog swimming activity, we calculated the average number of dog entries over 30 min observation periods in each week and each pond. There were a total of ten observation periods, five per day for the two days sampled in each week. Hence, the total observations from each period were summed and divided by ten.

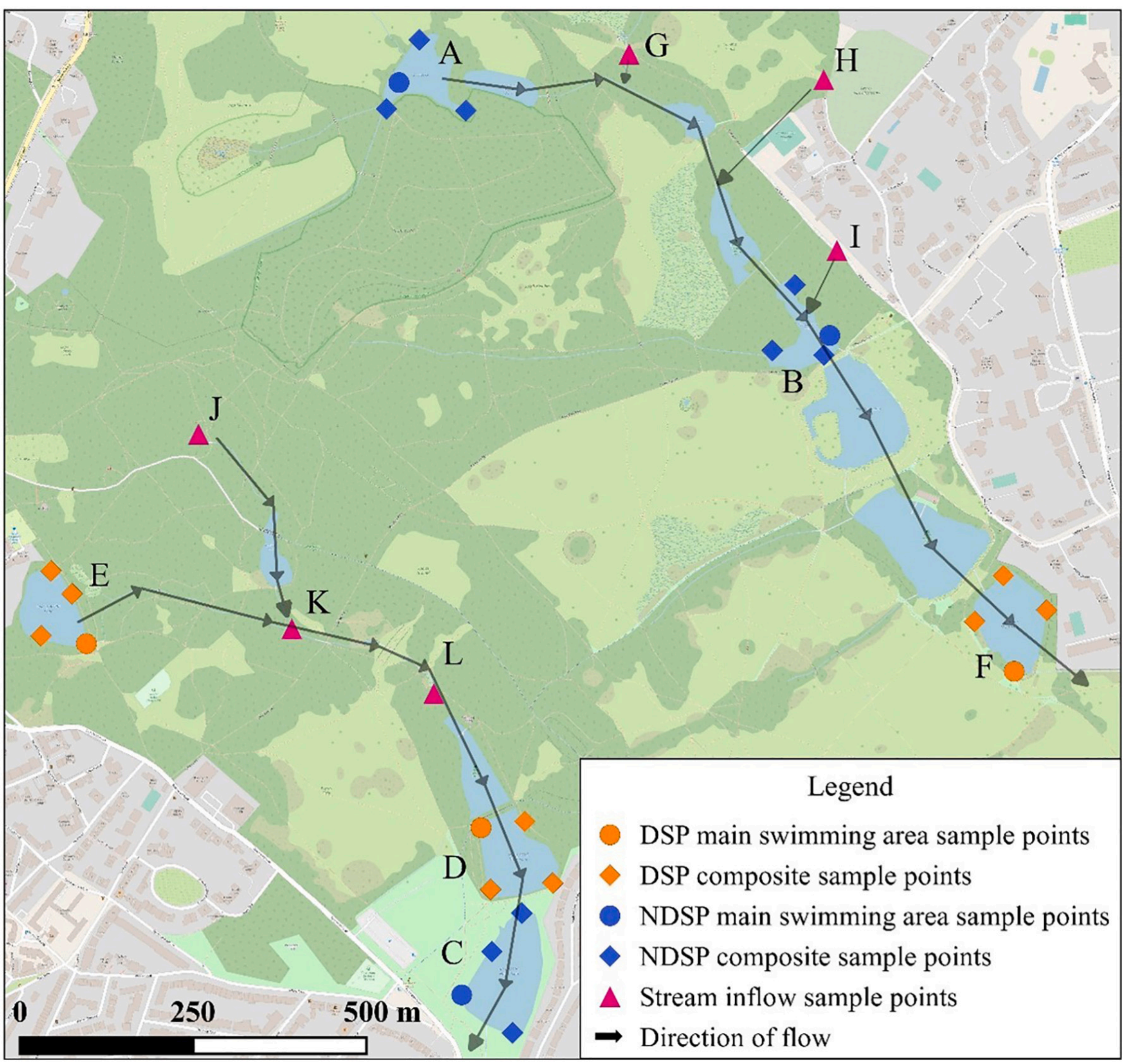

Fig. 1. Distribution of water sampling and dog observation points on Hampstead Heath. Blue shapes indicate sampling points at non-dog swimming ponds (NDSPs) A, B and C; orange shapes indicate sampling points at dog swimming ponds (DSPs) D, E and F. Main dog swimming areas and the point where single samples were obtained are shown as circles. The points where composite samples were obtained are shown as diamonds. Pink triangles (letters G, H, I, J, K, and L) indicate where a stream inflow sample was obtained. Direction of water flow is indicated by the arrows.

2.3. Analysis of pond water samples

2.3.1. Water sampling procedures

Samples were taken in duplicate from each location on two different dates (22/06/2023 and 05/07/2023). Prior to sampling, 30 mL Nalgene® (polypropylene) screw-capped bottles (Fisher Scientific, Loughborough, UK) and 10 L food grade buckets were pre-washed three times each with methanol and then ultrapure water. Samples were taken as per Egli et al. (2023). Water samples were collected from each pond per sampling week, after the Wednesday and Saturday dog observations. These comprised a single grab sample, taken from the designated main swimming point (at DSPs) or points under camera observation (at NDSPs) and an additional composite of grab samples taken from three other points around each of the ponds (Fig. 1). The grab samples from these three points were combined in equal proportions and shaken in a larger 1 L bottle, then transferred to a 30 mL Nalgene bottle to create a composite sample for each pond. The grabs for the composite samples were taken from points at least 50 m away from the main swimming point. All samples were taken approximately five to seven meters away from the edge of the pond at a depth of 0.3–0.6 m. Briefly, and at each sampling location, buckets were cast into ponds three times to pre-wash them and to wash Nalgene bottles. The bucket was rinsed with pond water between each sample. Single samples were transferred from the bucket to 30 mL Nalgene bottles. The samples were immediately put into a cool box with ice packs for transport to the laboratory and subsequently stored in the dark at −20 °C until analysis (within two weeks) to minimise degradation. In addition to pond samples, several stream samples were taken. Sources of water in Heath ponds come largely from springs arising on the Heath from rainfall percolating through sandy layers. In a few parts of the Heath, streams also enter from surrounding residential areas. Fig. 1 shows sampling points for streams originating on (J) and off (G, H, I) the Heath, as well as the sampling points in a stream flowing between a DSP and a downstream pond (K, L).

2.3.2. Reagents

All reagents were of analytical grade unless otherwise stated. Methanol, isopropanol, acetonitrile and formic acid (>95 % v/v) were purchased from Sigma Aldrich (Steinheim, Germany). A Millipore Milli-Q water system (Bedford, MA, USA) was used to generate ultrapure water to a resistivity of 18.2 mΩ.cm. Reference materials (>99 %) for fipronil and imidacloprid were ordered from Sigma and used for quantitative analysis. On top of pet parasiticides, a set of 163 reference materials for pharmaceuticals ($n = 97$), pesticides ($n = 55$) and illicit drugs ($n = 11$) were used as per Egli et al. (2023) for quantification by matrix-matched calibration. Standards were prepared as mixes to concentrations of 0.1, 0.01 or 0.001 μg/mL in methanol and were stored at −20 °C in the dark for use.

2.3.3. Sample preparation

All samples were prepared from a single aliquot and analysed in triplicate, meaning that each sample was prepared once but injected in triplicate during analysis (instrument/measurement replicates). Pond water samples were thawed at room temperature, agitated, and placed on a flat surface to allow for sedimentation to occur. Following sedimentation, a pooled sample containing aliquots of equal volumes from all pond samples was created by pipetting 900 μL from each sample into a sterile 50 mL centrifuge tube (Sigma-Aldrich, Steinheim, Germany). The pooled sample was used to prepare a 16-point external matrix-matched calibration line (0–5000 ng/L) as well as quality controls (QCs) at 250 and 2500 ng/L. Calibrants and samples were prepared by combining nine parts of the pooled sample or individual sample (900 μL), respectively with one part of diluent (100 μL) which consisted of a stable isotope-labelled internal standard mix of 36 compounds at 500 ng/L, and HPLC-grade methanol (Sigma-Aldrich, Dorset, UK) into 2 mL Safe-Lock Eppendorf tubes (Eppendorf, Hamburg, Germany). Additionally, calibrants were spiked with a standard mix of all other compounds resulting in each sample, calibrant and QCs containing a final volume of 1 mL. All samples were mixed using a vortex and centrifuged at 9000 rpm for 5 min at 15 °C. Samples were transferred into 750 μL centrifuge filter tubes (0.2 μM polytetrafluoroethane-based (PTFE) mesh (Thermo Scientific, Rockwood, USA)), mixed by vortex, and centrifuged at 9000 rpm for 5 min (centrifuge 5810 R, Eppendorf, Hamburg, Germany). Finally, the filtrate was transferred into 1.5 mL silanised amber glass LC-vials and capped (Agilent Technologies, Santa Clara, CA, USA) before immediate subsequent LC-MS analysis.

Quality controls were analysed at least twice throughout the sequence for each concentration. Signal intensity was checked across batches and assessed as acceptable if ≥90 % by the end of the run. Instrumental blanks were injected between samples, as well as in between QCs and calibrants to ensure no carryover between samples. At the beginning and the end of the sequence, tests were conducted to check for mobile phase contamination and shifts in retention time.

2.3.4. Instrumental analysis

Compound identification and quantification was performed using rapid direct-injection liquid chromatography tandem mass spectrometry (LC-MS/MS) according to Ng et al. (2020). Briefly, analytical separation was achieved in 5.5 min with a Raptor biphenyl guard column of 5.0 mm × 3.0 mm and 2.7 μm particle size (Thames Restek, Saunderton, UK) fitted to a Shimadzu Nexera X2 ultra high-pressure LC apparatus (Shimadzu Corporation, Kyoto, Japan). The injection volume was 10 μL and the flow rate 0.5 mL/min. Compounds were separated using the following elution program: an initial hold for 0.2 min of 10 % mobile phase B (MPB, 0.1 % v/v formic acid in 50:50 MeOH:MeCN; MPA = 0.1 % aqueous (v/v) formic acid), increase to 60 % MPB over 2.8 min, a hold at 100 % MPB for 1 min, then a re-equilibration period at initial conditions for 1.5 min. For mass spectrometry, a LCMS 8060 triple quadrupole mass spectrometer was used which was fitted with an electrospray source capable of switching polarity (Shimadzu Corporation, Kyoto, Japan). Multiple reaction monitoring (MRM) allowed confirmation of the detected analytes with two transitions.

2.3.5. Statistical analysis

Statistical analysis was done using Microsoft Excel (v. 2307) (Microsoft Corporation, Redmond, WA, USA, 2018) and R Statistical Software (v. 4.2.2) (R Core Team, Vienna, Austria, 2022). An analysis of covariance (ANCOVA) was used to estimate whether chemical concentrations (ng/L, in composite samples) were associated with the observed dog activity (i.e., total entries, which includes both entering the water and fully immersing) in the ponds (mean number of dogs/30 min recorded during the 10 counts: 5 per day x 2 days for each pond in each week of sampling). Compounds that were below the limit of quantification (<LLOQ) were marked as zero in calculations. The measure of average dog entries reflects a pattern of general usage for each pond in each sampling period, and with this in mind, the levels of imidacloprid and fipronil in the associated composite sample were used as the response variables. The composite samples were assumed to reflect accumulating chemical concentrations in the ponds from dog activity over time, whereas the main swimming point samples may reflect more variable and recent levels of chemical introduction. To account for there being two samples per pond, one from each sample week, 'week' was also included in the model as a two-level factor.

A paired *t*-test was used to compare the chemical concentrations recorded at the main dog swimming points with those of the composite samples taken from three other, more distant points at the same ponds. In the t-test, samples were paired by pond and day of sampling.

2.3.6. Environmental risk assessment

The environmental quality standard (EQS) data were taken from several sources to represent a range of values typically used in environmental policy and research. Sources included the EU biocides assessment report for fipronil, a European Commission proposal on EQSs in water policy, and a review of neonicotinoid toxicity studies, and the NORMAN Ecotoxicology Database, a network of reference laboratories, research centres and related organisations for monitoring of emerging environmental substances (ECHA, 2011; European Commission, 2022; Morrissey et al., 2015; NORMAN, 2023). For risk classification, risk quotients (RQ) were calculated using the highest measured concentration at each site (A-K) and divided by the lowest NORMAN PNEC value for freshwater (13 and 0.77 ng/L for imidacloprid and fipronil, respectively). Thresholds were set as per Palma et al. (2014), where high environmental risk was defined as RQ $\geq$ 10.0, medium risk as 1.0 $\leq$ RQ $<$ 10.0, low risk as 0.1 $\leq$ RQ $<$ 1.0, and insignificant risk as $<$0.1.

2.4. Questionnaire

The online questionnaire comprised 28 questions covering demographic information, time spent on Hampstead Heath, types of flea and tick treatments that owners use, factors that influence that use, and existing knowledge on any environmental risks of flea and tick treatments. Most answers were multiple choice, and a few were short answers (free text). Veterinarians, policymakers, environmental scientists, ecologists, social scientists, and chemists advised on the design of the questionnaire, including feedback on topics and wording of questions. The survey was checked for understanding by two non-experts. The questionnaire can be found in the Supplementary material.

2.4.1. Data collection

The questionnaire was designed and distributed using Jisc Online Surveys, a GDPR-compliant program. It was available from June to August 2023. To ensure that the responses were specific to dog owners who walked their dog on Hampstead Heath, the Heath & Hampstead Society assisted in facilitating the engagement of dog owners and distributing questionnaires through its membership and contacts with other Heath-focused charities and social media. Methods of distribution included newsletters, social media, websites, and signage. Dog owners were also approached while they were swimming their dogs to ask if they would complete the questionnaire.

All responses to the questionnaire were anonymous, consent to participate was obtained, and all respondents were over the age of 18. Respondents also certified that their answers related to a dog in their household that was walked on Hampstead Heath.

2.4.2. Data analysis

Statistical analysis was performed using Microsoft Excel (v. 2307) (Microsoft Corporation, Redmond, WA, USA, 2018) and SPSS (v. 29.0.1.0) (IBM Corp, Armonk, NY, USA, 2023). All variables were categorical and were presented by number and frequency.

2.4.3. Ethical approval

This study received ethical approval from the London School of Hygiene and Tropical Medicine MSc Research Ethics Committee (reference number: 28827).

3. Results

3.1. Dog activity and chemical concentrations

3.1.1. Sources of veterinary ectoparasiticides in the Hampstead Heath ponds

This study examined dog swimming on Hampstead Heath as a source of veterinary ectoparasiticides in its water bodies, but other sources were also considered. Imidacloprid and fiproles have been frequently measured in both raw and treated wastewater (Perkins et al., 2024; Teerlink et al., 2017; UKWIR, 2023), wastewater-impacted river waters in London (Egli et al., 2023, Richardson et al., 2022) and across the UK (Perkins et al., 2021). Any influx of wastewater into ponds could introduce these substances from surrounding urban areas, arising from down-the-drain disposal of flea treatment products, a pathway established by Teerlink et al. (2017) and Perkins et al. (2024). Although the Hampstead Heath pond catchment is largely isolated from urban areas and not known to be affected by local sewage and runoff, the potential for wastewater contributions through misconnected or 'leaky' sewer pipes to ectoparasiticide concentrations had to be excluded. Therefore, all water samples were profiled for 163 other chemicals of emerging concern (in addition to imidacloprid and fipronil) for evidence of contamination from wastewater sources. Generally, very few compounds were detected (and these were also detected infrequently) in the ponds (n = 14) compared to the 98 compounds detected in a recent study of 14 different waterways in London (Egli et al., 2023). Of these, seven (benzoylecgonine, carbamazepine, citalopram, cocaine, ketamine, methamphetamine, and warfarin) were quantifiable in Heath ponds and in these instances the average concentration was 19 $\pm$ 13 ng/L, which was similar (22 $\pm$ 13 ng/L) to that of the same compounds quantified in regions upstream of known CSOs and WWTP outfalls across four London rivers (R. Brent, R. Hogsmill, R. Lea, and R. Wandle; Fig. S1, Egli et al., 2023). The average concentrations of these seven contaminants in wastewater-impacted regions of the London rivers was roughly five-fold greater than those in the Heath ponds (19 $\pm$ 13 ng/L compared to 93 $\pm$ 94 ng/L, Fig. S1, Egli et al., 2023). While illegal drug use and direct human urination/disposal of waste material may have resulted in minor contamination of Heath ponds with chemical contaminants, this was considered a very minor and infrequently detectable contribution.

In addition, samples from streams fed by a spring on the Heath and leading into ponds were negative for both imidacloprid and fipronil (Point J in Fig. 1), as well as for samples from streams originating from residential areas on the edges of the Heath (Points H and I in Fig. 1). These results indicate that the sources of imidacloprid and fipronil in Heath ponds were more likely to derive from animals in pond waters rather than an external source such as wastewater contamination.

3.1.2. Comparison of veterinary ectoparasiticide concentrations across ponds

Dog swimming activity in different ponds is shown in Table 1. The majority, 68 % - 98 %, of dog entries observed in DSPs were at the main swimming point, though some dogs took alternative routes to water. Dog immersions ranged between 36 % and 65 % of total dog entries in different DSPs on different dates. Complete dog activity data is presented in Supplementary Data 1.

An ANCOVA including average dog entries per 30 min and week of recording as factors to account for chemical concentrations in composite samples was run twice: once including the NDSPs which provide anchoring 0,0 values, and once without. Fig. 2 is a graphical representation of this analysis, with the solid lines representing the relationship between average dog entries and chemical concentrations for all ponds, and dashed lines for DSPs only.

For imidacloprid, with all ponds included, the model accounted for much of the variance in these data (F = 46.03, d.f. = 2,9, $p < 0.001$, R^2 = 0.91) with average dog entries accounting for the majority of this (R^2 = 0.89). With the NDSPs excluded from the model, the explanatory power of average dog entries fell (R^2 = 0.72) but remained a detectable influence (F = 14.71, d.f. = 1,3, p < 0.00514) and the correlation remained similar.

The picture was similar, though less clear, for fipronil as overall model fit was poorer. With all ponds included, the model accounted for 75 % of the variance in these data (F = 17.13, d.f. = 2,9, $p < 0.001$, R^2 = 0.75) with average dog entries accounting for the majority of this (R^2 = 0.69). With the NDSPs excluded from the model the explanatory power of average dog entries fell (R^2 = 0.19) and any influence was not detectable (F = 2.83, d.f. = 1,3, p = 0.19).

To examine the environmental risk associated with the concentrations observed, lowest predicted no effect concentrations (PNECs) for these compounds were obtained from the literature and compared to

Table 1
Dog activity (entry and immersion) and ectoparasiticide concentrations taken at the main swimming point and a composite measure of three further points within each pond at a June and July sampling date (±SD). Samples from streams were taken in July. Full results are presented in Supplementary Data 1 and 2. <LLOQ – detected, but below the lower limit of quantitation (limit of detection [LOD] = 2.0 ng/L [fipronil] and 1.9 ng/L [imidacloprid]; LLOQ = 5.7 ng/L [fipronil] and 6.0 ng/L [imidacloprid]).

Location	Sample week	Dog entry[a]		Dog immersion[a]		Imidacloprid (ng/L)		Fipronil (ng/L)	
		Main swim point	All entries	Main swim point	All immersions	Main swim point	Composite	Main swim point	Composite
Non dog swimming ponds									
A	1	0.1 ± 0.3	0.1 ± 0.3	0.1 ± 0.3	0.1 ± 0.3	<LOD	<LOD	<LOD	<LOD
	2	0.0	0.0	0.0	0.0	<LOD	<LOD	<LOD	<LOD
B	1	0.0	0.0	0.0	0.0	<LOD	<LOD	<LOD	<LOD
	2	0.0	0.0	0.0	0.0	<LOD	<LOD	<LOD	<LOD
C	1	0.0	0.0	0.0	0.0	<LOD	<LOD	<LOD	<LLOQ
	2	0.0	0.0	0.0	0.0	<LOD	<LOD	<LOD	<LOD
Dog swimming ponds									
D	1	6.8 ± 5.4	8.9 ± 6.7	2.9 ± 2.6	4.2 ± 3.4	121.0 ± 7.0	73.0 ± 7.0	16 ± 0 ± 3.0	7.0 ± 1.0
	2	3.1 ± 2.6	5.6 ± 3.3	0.6 ± 1.1	2.0 ± 1.9	54.0 ± 3.0	<LOD	<LLOQ	5.0 ± 1.0
E	1	19.1 ± 9.4	19.6 ± 9.7	11.0 ± 5.4	11.2 ± 5.5	784.0 ± 10.0	152.0 ± 7.0	85.0 ± 10.0	10.0 ± 2.0
	2	11.6 ± 6.0	11.8 ± 6.1	4.8 ± 3.2	4.9 ± 3.1	280.0 ± 4.0	157.0 ± 3.0	26.0 ± 5.0	19.0 ± 5.0
F	1	22.3 ± 7.0	24.2 ± 7.9	14.5 ± 4.5	14.6 ± 4.7	317.0 ± 7.0	208.0 ± 8.0	50.0 ± 10.0	13.0 ± 6.0
	2	15.0 ± 8.4	17.2 ± 9.8	7.8 ± 4.8	8.1 ± 5.2	298.0 ± 4.0	210.0 ± 2.0	16.0 ± 3.0	15.0 ± 5.0
Streams									
G	2	N/A		N/A		<LOD		<LOD	
H	2	N/A		N/A		<LOD		<LOD	
I	2	N/A		N/A		<LOD		<LOD	
J	2	N/A		N/A		<LOD		<LOD	
K	2	N/A		N/A		201.0 ± 2.0		12.0 ± 7.0	
L	2	N/A		N/A		35.0 ± 5.0		<LOD	

[a] Mean of 10 observations of 30 min each.

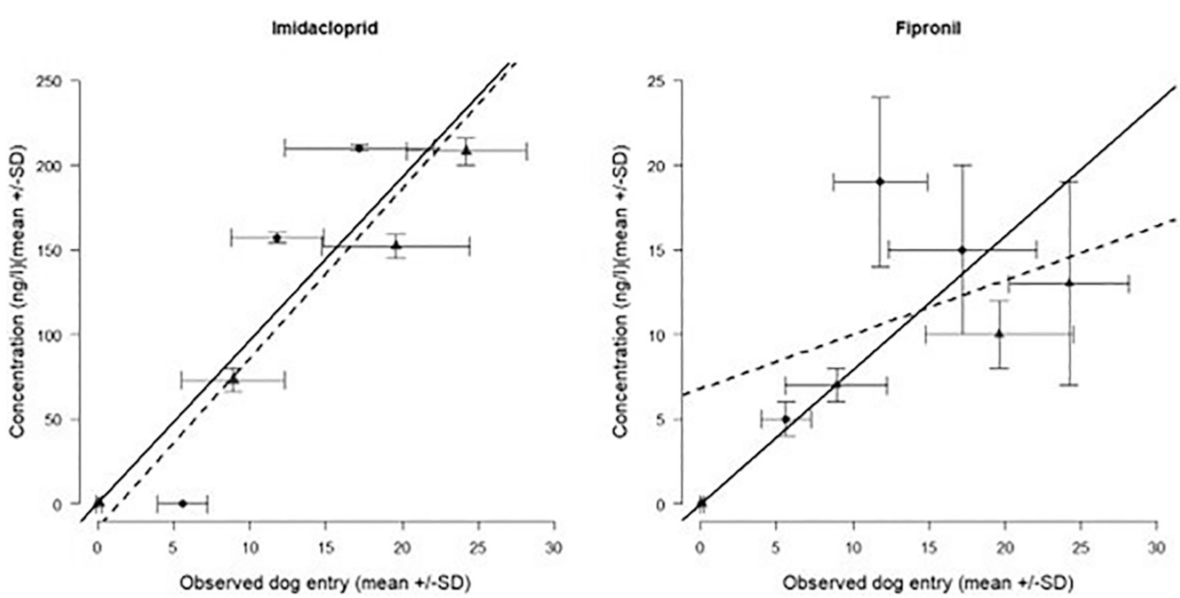

Fig. 2. Concentrations (±SD) of imidacloprid (L) and fipronil (R) in composite samples as a function of observed dog entries (±SD) (dogs entering the pond per 30 min) at those points for each pond. Solid lines indicate the relationship including NDSPs and DSPs, and dashed lines where only DSPs are included.

levels found in DSPs. For 11 out of 12 samples taken in DSPs, concentrations in samples taken in both the main and composite sample points exceeded the NORMAN PNECs for imidacloprid (13 ng/L) and fipronil (0.77 ng/L). The PNEC values of the fipronil breakdown products range from 12.0 to 35.0 ng/L, indicating that they pose a lower risk to the environment than fipronil (NORMAN, 2023). Fig. 3 shows these results in graphical form, where sample parasiticide concentrations were averaged for the different dates for a particular pond and sample. It can be seen in Fig. 3 that most DSP samples exceeded not only NORMAN PNECs, but other established freshwater environmental quality standards such as the European Commission (6.8 ng/L for imidacloprid) and the European Chemical agency (12.1 ng/L for fipronil) (European Commission, 2022; ECHA, 2011). Most DSP samples of imidacloprid exceeded the chronic toxicity limit (35 ng/L) proposed by Morrissey et al. (2015), and two exceeded the acute toxicity limit (200 ng/L).

Risk quotients (RQ) were calculated for concentrations in main and composite samples at each of the three DSPs. For imidacloprid, quotients indicated medium risk ($RQ_{main} = 9$; $RQ_{composite} = 6$) for pond D and high risk for the other two DSPs ($RQ_{main} = 60$ and $RQ_{composite} = 12$ for pond E; $RQ_{main} = 24$ and $RQ_{composite} = 16$ for pond F). For fipronil, all DSPs were

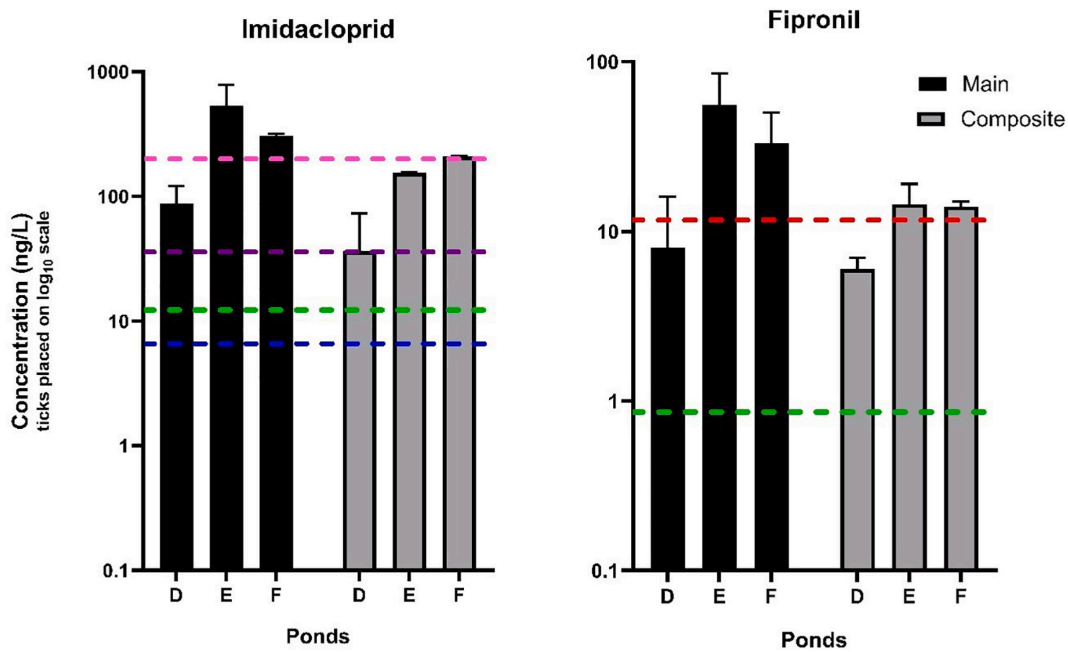

Fig. 3. Mean (+maximum, as whiskers) concentrations (ng/L) of imidacloprid and fipronil concentrations from the 12 samples taken at dog swimming ponds on Hampstead Heath in the summer of 2023 plotted on a $\log_{10}$ y-axis. The dotted lines indicate the values of the environmental quality standards as defined by the EU, NORMAN, and Morrissey et al. (2015). The green line marks the lowest freshwater predicted no effect concentration (PNEC) of 13 ng/L for imidacloprid and 0.77 ng/L for fipronil (NORMAN, 2023). Note that the LOQ for fipronil was 5.7 ng/L. The blue line is the AA-EQS (average annual – environmental quality standard) for imidacloprid of 6.8 ng/L (European Commission, 2022), and the red line is the PNEC for freshwater of 12.1 ng/L from the 2011 EU assessment report for fipronil (ECHA, 2011). The pink line represents the acute toxicity limit of 200 ng/L and the purple line represents the chronic toxicity limit of 35 ng/L for imidacloprid as presented by Morrissey et al. (2015).

at high risk at all ponds for main swimming points (RQ = 21, 110 and 65 for ponds D, E and F, respectively) and composite points (RQ =10, 24, and 19 for ponds D, E, and F, respectively).

3.1.3. Dispersion of veterinary ectoparasiticides in the water system

Main dog swimming points are likely to show the highest, and most variable, concentration of imidacloprid and fipronil in a particular pond, as they will be substantially influenced by recent activity. These chemicals, though, do have capacity to disperse throughout a pond and the presence of the compounds in composite samples indicates that dispersion does happen. In DSPs, the observed average concentrations of chemicals were indeed higher at main swimming points for imidacloprid (309 ± 104 vs 133 ± 34 ng/L) and fipronil (32 ± 13 vs 12 ± 5 ng/L). However, there was no evidence in this small sample that the mean concentration of either chemical varied systematically between the main dog swimming point and the composite sample from three other places around the pond (imidacloprid: $t = 1.71$, df = 11, $p = 0.11$; fipronil: $t = 1.55$, df = 11, $p = 0.15$). The samples, however, did have substantially different variance patterns (imidacloprid: $F_{11,11} = 7.04$, $p = 0.003$, fipronil: $F_{11,11} = 14.67$, $p < 0.001$) suggesting that the composite samples provide a more consistent representation of pollution from these compounds than do samples taken at the main entry points which may reflect strongly the most recent entries and thus mis-represent general levels. However, samples taken at main entry points provide measures for maximum concentrations and therefore maximum exposure levels in the ponds which can be compared to risk thresholds (Fig. 3).

Further information on ectoparasiticide dispersion was available from samples taken between and below DSPs. Two samples were taken from an intermittent stream below Pond E, (Points K and L in Fig. 1). These samples included outflow from another pond east of Pond E where dog swimming also occurs. Point K, which was more proximate to these DSPs, recorded measurable levels of both imidacloprid (201 ng/L) and fipronil (12 ng/L), while a Point L further downstream recorded only imidacloprid (35 ng/L). Finally, Pond C, an NDSP, is supplied by outflow from Pond D, a DSP, but no ectoparasiticides were recorded there (see Table 1).

3.2. Questionnaire results

The questionnaire was directed at a specific community of dog owners who walk and swim their dogs on Hampstead Heath. While the sample was small, it provided useful information on the likely origin of the veterinary ectoparasiticides found in ponds and on the treatment-related practices and beliefs of people who swim their dogs there. Full questionnaire results are presented in Supplementary Data 3.

Out of the 108 submitted questionnaires, seven were removed due to incomplete responses, leaving 101 questionnaires for analysis. Of the 101 people who completed the questionnaire, 81 % swam their dogs on the Heath. Most of those who swam their dogs did this daily (30 %) or weekly (43 %). Amongst the 101 dog owners, patterns of treatment with flea and tick products varied widely. Most respondents (78 %) indicated that they treated their dog regularly whether fleas or ticks were present or not, while 9 % indicated that they treated their dog in response to the presence of fleas or ticks, 7 % of respondents stated that they did not treat, and 6 % used other methods. When the 87 respondents who used flea treatments in the past 12 months were asked to estimate how often they treated their dogs, 33 % said they treated their dog 12 times, 21 % treated 6–11 times, 42 % treated 1–5 times, and 3 % treated >12 times. Most of the 101 respondents (70 %) did not report receiving any information about how long they should wait after treatment before allowing their dog to swim. The 30 respondents that did, received this information largely from veterinarians (70 %; $n = 21$) and packaging (53 %; $n = 16$).

Amongst the 87 respondents who used flea treatments in the past 12 months, there was considerable variation in the type of products used, and therefore in the active ingredients and mode of application. All were able to identify the mode of application of their flea and tick treatments. Over half used products delivered as pills or tablets (59 %), and a substantial proportion (46 %) used spot-on, collar, or spray-on products, which included products containing imidacloprid and fipronil. Most (69 %) of these 87 respondents were able to identify the products they had used in the past 12 months from a provided list of UK products. Of the 60 who could identify which products they had used, around half reported using flea treatments containing imidacloprid or fipronil, (28 % and 22 % respectively). Out of the 27 respondents who used a product containing imidacloprid and/or fipronil, 7 % (n = 2) had received information on the environmental harms posed by flea treatments, and 48 % (*n* = 13) had received advice to wait following application before allowing their dog to swim.

Almost two-thirds (63 %) of the 68 respondents who used flea treatments in the past 12 months and swam their dogs were able to identify the products they used, and of these 44 % (*n* = 19) used a product containing imidacloprid and/or fipronil. Additionally, 13 % (*n* = 9) of respondents who swam their dogs and used flea treatment had received information on the environmental harms posed by flea treatments, and 34 % (*n* = 23) had received information on waiting between applying treatment and allowing their dog to swim. Most (90 %) identified veterinarians as their source of information on flea and tick treatments. Other sources mentioned were online information (29 %) and product packaging (19 %).

Most (86 %) of the 101 respondents were unaware of the potential risks of flea treatments to aquatic wildlife and environments. When asked how important protecting nature would be to their decision on what product to use and how it should be used, most (67 %) said that it would be very important, 27 % said that it was fairly important, and 6 % said it was not important.

4. Discussion

This study demonstrates that dog swimming on Hampstead Heath was highly likely to be responsible for the introduction of the veterinary ectoparasiticides imidacloprid and fipronil into ponds. Measurable concentrations of both chemicals were only found in ponds with dog swimming (DSPs) and in streams below these ponds. A highly significant association was found across all ponds between a measure of dog entries into ponds and composite concentrations of both chemicals. Eliminating the NDSPs, where no quantifiable levels of these chemicals were found, this significant association remained for the three DSPs for imidacloprid, but not for fipronil.

The study suggests that, for DSPs, points at which dogs regularly enter ponds exhibit particularly high and variable levels of both chemicals. This variability may reflect the fluctuation of ectoparasiticide concentrations in the fur of dogs swimming at particular times, as the questionnaire revealed that around a quarter (28 %) of dogs swimming were treated with products containing imidacloprid or fipronil in the past 12 months. The lower variability in concentrations of these chemicals in composite samples, taken around pond edges distant from main swimming points, supports our presumption that these samples give a better indication of persistent and chronic levels of ectoparasiticides in ponds. Dog swimming is a daily and year-round activity on Hampstead Heath, but it is possible that variations in ectoparasiticide levels may occur in response to seasonal changes in dog activity, rainfall and water flow in ponds.

One potential limitation of this study is the choice of the parent compound fipronil, as it undergoes photodegradation in water to desthiofipronil with fipronil amide, -sulfide, -sulfone, also known to form as minor breakdown products (Ngim and Crosby, 2009; Singh et al., 2021). Desthiofipronil is more persistent than the parent compound in water and could yield a better correlation with dog activity in the ponds. Unfortunately, this compound was not included in the targeted LC-MS/MS method. However, other studies have shown that across several matrices, the parent compound fipronil has higher detection frequencies compared to its transformation products (Perkins et al., 2021, Cryder et al., 2019) and with a comparatively long half-life is a reasonable choice of analyte for assessing the risks of ectoparasiticides in the environment arising from dog swimming.

Almost all of the imidacloprid and fipronil concentrations in DSPs exceeded a range of established measures for environmental toxicity thresholds. On this basis, we conclude that their presence poses a risk to aquatic biodiversity. A range of invertebrate taxa have been shown, in macrocosm and/or field studies, to be negatively impacted by imidacloprid and fipronil, including aquatic insects (Pisa et al., 2015; Yamamuro et al., 2019; Barmentlo et al., 2019; Miller et al., 2020; Schmidt et al., 2022). Further work on Hampstead Heath should assess differences in invertebrate diversity and abundance between ponds with and without dog swimming. As Hampstead Heath is a protected, Metropolitan Site of Interest for Nature Conservation, studies might focus on impacts that affect charismatic and protected species. For instance, the Heath has an unusually rich community of odonates for an urban setting (Andrew, 2022). Other taxa of flying aquatic insects support local populations of London Priority Species (Greater London Assembly, 2019), including Swift (*Apus apus*) and Water Bat (*Myotis daubentoni*). A reduction in invertebrates that support fish could impact on a local angling community and on the performance of the Heath's breeding population of Kingfisher (*Alcedo atthis*), another London Priority Species.

While high concentrations of imidacloprid and fipronil were found in some Heath ponds, we think it unlikely that Hampstead Heath is a significant source of these chemicals in waters flowing from the Heath relative to other domestic ectoparasiticide sources affecting those waterways. Both chemicals were present in some but not all samples from streams and ponds between and below DSPs. Stream sampling site K downstream from site E had high risk quotients of 15 and 7 for imidacloprid and fipronil, but we note that another pond with dog swimming, visible on Fig. 1, also feeds into this stream. Less than 250 m downstream from this point, at site L, risks were reduced to moderate for imidacloprid (RQ = 3) and fipronil was not detected. The only risk area of ectoparasiticide release from the ponds studied into wider waterways was site F. Assuming the same degree of dilution as site L, it is expected that risks would be reduced over a similar distance. By contrast, consistently high concentrations of imidacloprid found across London waterways are associated with both treated and untreated municipal wastewater sources (Perkins et al., 2024; Egli et al., 2023; Vane et al., 2022; Richardson et al., 2022; Munro et al., 2019; White et al., 2019). In a study of South London's River Wandle in 2021, for instance, imidacloprid was only quantifiable in river water samples collected at and downstream of the wastewater treatment plant, and was not detected above the outfall, even in river water samples collected from popular parks for dog walking and swimming (Richardson et al., 2022).

This study shows that restricting dog swimming may be an effective way to reduce concentrations of ectoparasiticides in ponds. Results from Pond D (Table 1) suggest, albeit not under controlled conditions, that dog exclusion may lead to rapid change in concentrations. At this DSP, between the first and second sample date, signage was installed to caution dog owners about high levels of harmful, blue-green algae. The subsequent sample, two weeks later, revealed a sharp decline at the main swimming point in dog activity (6.8 to 3.1 entries per 30 min) and in concentrations of imidacloprid (121 to 54 ng/L) and fipronil (16 to below quantifiable concentrations).

A number of publications for dog owners identify measures that may reduce risks of ectoparasiticide pollution, such as Safe-Dog-Swimming (BVA, BSAVA, and BVZS, 2021; Veterinary Prescriber, 2023). Recommended actions from Veterinary Prescriber (2023) include not swimming dogs for a period after treatment with spot-on products; treating dogs on a risk-based, rather than a regular, prophylactic basis; and using products that do not contain topically applied ectoparasiticides that wash off dogs in water.

Responses to the questionnaire revealed opportunities to encourage all of these practices in a local context. Answers to questions about both products and mode of application used revealed that about a third of owners of swimming dogs who used flea and tick treatments were not aware of recommendations to wait between treating and swimming their dogs, and very few were aware of their environmental risks. However, nearly all of the respondents indicated that protecting nature would be important in their decision-making on the type and method of flea treatment used.

Prophylactic treatment with ectoparasiticides has grown rapidly in the UK in recent years, despite strong arguments for a risk-based approach to reduce unnecessary environmental pollution (Whitehead and Perkins, 2022; Farrell et al., 2023). Dog owners visiting Hampstead Heath follow this national trend, but it is noteworthy that around 20 % are already treating their dogs on an "at need" basis or not using flea or tick treatments at all, a proportion that could perhaps be grown through improved information to dog owners on how to protect their dogs without using more flea or tick treatment than necessary.

Finally, while around half of dog owners in our study who reported using flea treatments in the past 12 months used spot-on, collar, or spray-on products that are likely to contain imidacloprid and/or fipronil, use of other products was also reported. Most of these were tablet- or pill-based products containing systemically-active isoxazoline insecticides. The residues of such products are excreted primarily in faeces or urine, but more research is needed to determine whether this would lead to less pond contamination in comparison to spot-on products and collars. Environmental pathways and risks associated with these systemic products remain poorly known (EMA, 2022). The important point from our study is that there is great diversity of products being used, and hence more opportunity exists for environmentally friendly products to compete with alternatives.

This study found that dog owners who swim their dogs on Hampstead Heath relied predominantly on veterinarians for information and guidance on the use of flea and tick treatments. We suggest therefore that local veterinary practices be closely involved with managers of urban green spaces and dog owners in action to reduce local environmental risks posed by veterinary parasiticides.

5. Conclusions

The veterinary ectoparasiticides imidacloprid and fipronil have been measured at concerningly high concentrations in ponds at an urban green space where large numbers of dogs are allowed to swim. Their absence from nearby ponds where dogs are excluded, and the absence of chemical indicators suggesting other wastewater sources, confirmed that dog swimming was the most likely source. Concentrations of these chemicals at points where dogs enter ponds ranged from 54 to 784 ng/L for imidacloprid and from below the limit of quantitation to 85 ng/L for fipronil. In all three of these dog swimming ponds, concentrations of both products at sampling points distant from these main swimming points exceeded environmental "predicted no effect concentrations" and exhibited medium to high risk quotients. A survey of people who swim their dogs on Hampstead Heath revealed that 86 % were unaware of the environmental risks posed by veterinary ectoparasiticides, but that 94 % considered protecting nature would be important to their choice of products. Besides the positive effect of excluding dogs from ponds, current treatment practices by dog owners identified opportunities for reducing contamination by these chemicals, including using other parasiticide products, reducing frequency of treatment by adopting a risk-based approach and following recommendations on delaying swimming after treatment. Urban green spaces are key targets for ectoparasiticide risk reduction, due to the value placed on their biodiversity and their high levels of dog activity. Locally, we recommend a multi-stakeholder approach to adopting risk reduction strategies involving green space managers, veterinarians, and dog owners, while nationally, we suggest that industry and regulators have a complementary role to play in improving the safety of products available for use.

CRediT authorship contribution statement

Lauren E. Yoder: Writing – review & editing, Writing – original draft, Methodology, Investigation, Conceptualization. **Melanie Egli:** Writing – review & editing, Validation, Software, Methodology, Investigation, Formal analysis, Data curation. **Alexandra K. Richardson:** Writing – review & editing, Validation, Methodology, Investigation, Formal analysis. **Adrian Brooker:** Writing – review & editing, Investigation, Conceptualization. **Rosemary Perkins:** Writing – review & editing, Visualization, Formal analysis. **C.M. Tilly Collins:** Writing – review & editing, Visualization, Formal analysis. **Jacqueline M. Cardwell:** Writing – review & editing, Supervision, Methodology, Conceptualization. **Leon P. Barron:** Writing – review & editing, Writing – original draft, Supervision, Resources, Project administration, Methodology, Funding acquisition, Data curation, Conceptualization. **Jeff Waage:** Writing – review & editing, Writing – original draft, Supervision, Project administration, Methodology, Investigation, Conceptualization.

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.

Data availability

All data used in the project is available in the attached Supplementary Materials files 1, 2 and 3 in Appendix A.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2024.176686.

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