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Dead in the water: comment on "Development of an aquatic exposure assessment model for imidacloprid in sewage treatment plant discharges arising from use of veterinary medicinal products"

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Abstract

Anthe et al. (Environ Sci Eur 32:147, 2020. https://doi.org/10.1186/s12302-020-00424-4) develop a mathematical model to calculate the contribution of veterinary medicinal products (VMPs) to the levels of imidacloprid observed in the UK water monitoring programme. They find that VMPs make only a very small contribution to measured pollution levels, and that the estimated concentrations do not exceed ecotoxicological thresholds. However, shortcomings in methodology—including the implicit assumption that imidacloprid applied to pets is available for release to the environment for 24 h only and failure to incorporate site-specific sewage effluent data relating to measured levels—raise questions about their conclusions. Adjusting for these and other deficiencies, we find that their model appears consistent with the conclusion that emissions from VMPs may greatly exceed ecotoxicological thresholds and contribute substantially to imidacloprid waterway pollution in the UK. However, the model utilises imidacloprid emissions fractions for animals undergoing the different scenarios (for example, bathing) that are extrapolated from unpublished studies that do not clearly resemble the modelled scenarios, with insufficient evidence provided to support their derivation. As a result, we find that the model presented by Anthe et al. provides no reliable conclusions about the contribution of veterinary medicinal products to the levels of imidacloprid in UK waterways.

Keywords: Imidacloprid, Waterway, Pollution, Wastewater

Background

Imidacloprid has been found to contaminate many surface waters around the world, at levels that pose a significant risk to the diverse communities that these ecosystems support [10, 16, 21]. Recent studies have raised concerns that environmentally harmful quantities of imidacloprid used in topical flea products may be passing to waterways from treated pets [25, 27, 29]. Indeed, initial calculations of exposure concentrations in surface waters from the treatment of pets with imidacloprid in The Netherlands show that the environmental threshold of 8.3 ng/l would be exceeded if only 1.15% of applied imidacloprid passed from treated pets to waterways via household drains [19]. Teerlink et al. [31] demonstrated that washing 25% of treated dogs within one week of applying a spot-on product containing fipronil would account for the entire fiprole load seen in Californian sewersheds.

Anthe et al. [1] present a model, funded by Bayer, manufacturer of imidacloprid, that estimates imidacloprid levels in emissions from UK sewage

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treatment plants (STPs) resulting from the use of dog and cat spot-on and collar veterinary medicinal products (VMPs) containing imidacloprid, and thereby calculate a predicted environmental concentration (PEC) in waterways. Anthe et al.'s model estimates the amount of imidacloprid applied daily to a population of cats and dogs in the catchment of an STP serving a human population of 10,000, then estimates how much of that imidacloprid passes from treated pets via STPs to waterways per day, via three scenarios—bathing pets, washing pets' bedding, or walking in rain—based on the likelihood of each scenario occurring per day. Anthe et al. conclude that their model demonstrates that veterinary spot-on and collar products make only a very small contribution to the levels of imidacloprid observed in the UK water monitoring programme, and that the contribution from veterinary use does not exceed ecotoxicological threshold values. However, we argue that several of the assumptions underlying the model result in substantial underestimation of the contribution of veterinary flea products to the measured imidacloprid pollution of waterways, and that the model is based on unsubstantiated emissions fractions, calling into question the validity of their conclusions.

Critique of Anthe et al.'s model

The model does not account for imidacloprid's persistence on pets

The model assumes that the amount of imidacloprid on a population of pets available for release to an STP per day is equal to the amount applied to that population per day (their Eq. (4), our Fig. 1). This assumption is incorrect, because much of the imidacloprid applied to dogs and cats persists for at least 4 weeks [6, 20]. Therefore, the amount of imidacloprid on a population of pets available for release to the environment on any one day is far higher than the amount applied to the population on that day. From Anthe et al.'s Table 1, 81.7% of imidacloprid is applied to pets in spot-on products. Craig et al. [8] measured imidacloprid residues transferred onto gloves at 24 h, 72 h, then weekly intervals for 5 weeks following application of a spot-on. This study found that the transferrable residue declined with time, being at 72 h 40% of that at 24 h, and remained detectable for 4 weeks. By disregarding the imidacloprid remaining on pets after the day of application, Anthe et al.'s Eq. (4) excludes the bulk of imidacloprid available for release to the environment throughout the rest of their model. Furthermore, there is a lack of clarity surrounding the amount of imidacloprid

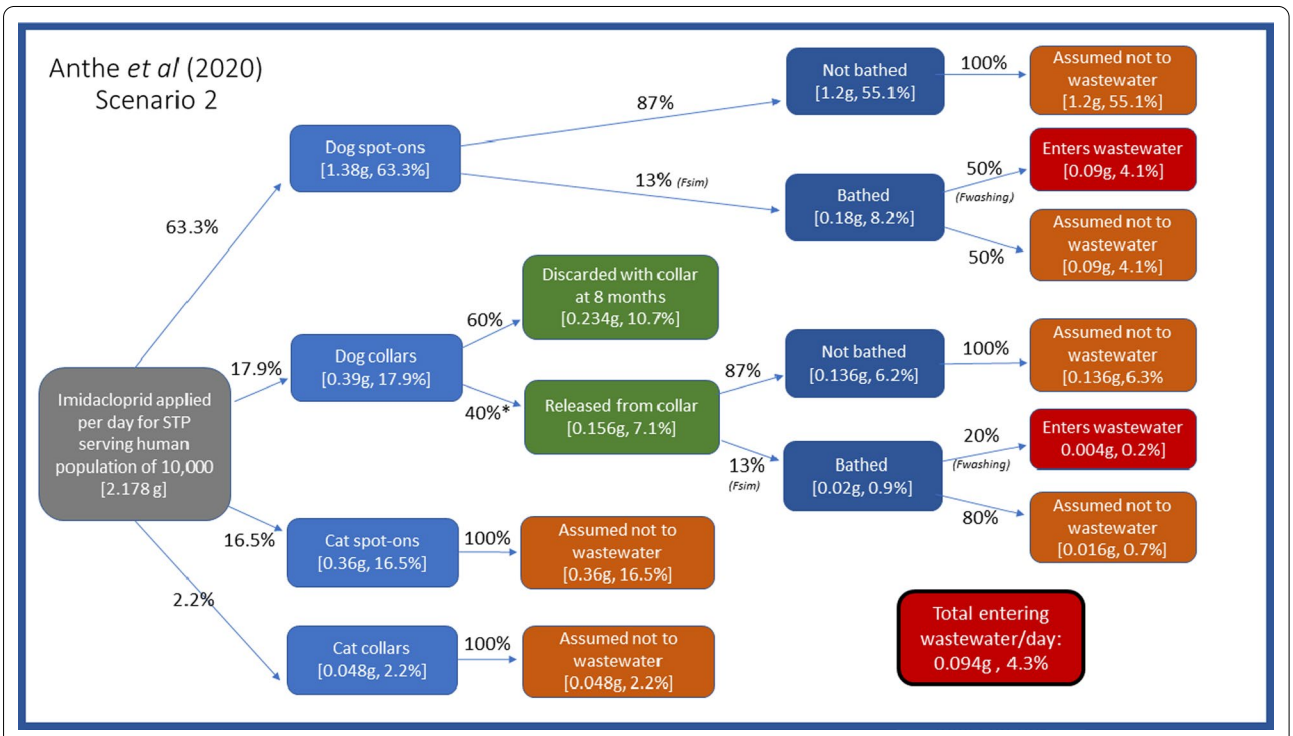

Fig. 1 Flow diagram depicting Scenario 2/the "worst case scenario" presented by Anthe et al. [1], illustrating the daily flow of imidacloprid, resulting from bathing dogs, through a standard STP (Sewage Treatment Plant) serving a human population of 10,000 people. Square brackets indicate amount of imidacloprid in grams and percentage of total imidacloprid applied per day. *Calculated from Anthe et al. Eq. (12) and Table 1. F_{sim} = fraction of dogs bathed per day (simultaneity factor), $F_{washing}$ = emission fraction of imidacloprid released due to bathing. F_{sim} and $F_{washing}$ from Anthe et al. [1]

assumed to be present on, and so available for release from, pets treated with collars. The authors calculate this amount by dividing the quantity of imidacloprid in collars by 240, that being the number of days registered duration of efficacy for such collars. However, no pharmacokinetic evidence is provided to support this approach, which appears to be based on the unsupported and improbable assumption that imidacloprid, once released from the collar, is present on the animal for only 24 h.

Persistence and accumulation of imidacloprid on pet bedding

In calculating the imidacloprid released through the washing of bedding, the authors do not appear to account for the fact that imidacloprid abraded off pets accumulates, and persists for months, on pet bedding [18]. The model assumes that a fixed proportion (Anthe et al.'s F_{abr}) of applied imidacloprid will abrade onto the pet's bedding, and so be available to be washed off. But F_{abr} is estimated for only the day that imidacloprid is applied to the pet, disregarding that the amount of imidacloprid abraded on to, and remaining on, pets' bedding will increase over the days, weeks or months since the bedding was last washed.

Disregarding additional pathways to waterways via STPs

In considering only three routes from treated pets to waterways—bathing dogs, washing pets' bedding, and walking in rain—the model disregards several other likely pathways for imidacloprid to STPs. Bigelow Dyk et al. [2] demonstrated the transfer of fipronil applied in spot-on flea products onto multiple surfaces and textiles within residential interiors, included items that are washed, such as family's hands and socks—providing evidence for the existence of pathways for substances in topical pet treatments to the sewage system not considered by Anthe et al. Imidacloprid is present in house dust [28, 30], with higher concentrations in households with pets [28], and some house dust may enter drains, e.g., during household cleaning. Additionally, a proportion of imidacloprid ingested by pets during grooming may be excreted in urine and faeces, based on studies on other species [14, 15, 17, 32]. Forster et al. [14] found that imidacloprid was one of the most frequently detected pesticides in the urine of dogs. Some urine and faeces from cats passes into the sewage system through flushing of cat litter [7], and surface runoff from closed surfaces is another potential route for contaminated excreta to STPs [9].

Discounting non-STP pathways to waterways

Anthe et al. do not consider non-STP pathways of imidacloprid from pets to waterways. Critically, Anthe et al.'s model disregards pets swimming in rivers as a possible source of pollution. Their justification being that swimming "is considered a sporadic and very localised incident, so emissions from this scenario are not pertinent to the monitoring data observed throughout the year under the WF WLD [Water Framework Directive Watch List] and nor are they pertinent to the developed model which focuses on emissions from STP". However, no evidence is provided to support the supposition that treated pets swimming in waterways does not contribute significantly to waterway pollution from imidacloprid, including in sites local to STPs, nor to support the assumption that this is less significant than the pathways included in the model. Further, Anthe et al.'s Eq. (12) and Table 1 reveal that 60% of the imidacloprid inventory in collars remains at 8 months. This will be disposed of in local waste, as are used spot-on pipettes containing residual imidacloprid, any unused spot-on pipettes, and imidacloprid-contaminated household dust entering vacuum cleaners. This will amount to several hundred kilogrammes per year of imidacloprid going to landfill—another potential source of waterway pollution, through leaching into groundwater and surface water from landfill [3].

Discounting concurrent pathways to waterways

The model assumes that if a fraction of applied imidacloprid passes from pets to waterways via one pathway, such as bathing, then no further imidacloprid will pass to waterways through other pathways, such as washing pet bedding. The authors state that "the model outputs from the different scenarios were not summed which recognises the interconnections between the scenarios and avoids double counting". However, if a portion of applied imidacloprid passes to waterways through one pathway, such as bathing a portion of pets, this does not preclude imidacloprid passing to waterways through other pathways from the pets that have not been bathed (constituting 87% of dogs on any one day, according to the model, see Fig. 1)—or even from pets that have been bathed, unless bathing removes all the applied imidacloprid. Imidacloprid may pass from pets to waterways via multiple pathways and Anthe et al.'s disregard for this further underestimates the imidacloprid originating from pets in their model.

Emissions from treated cats

Two of the three scenarios presented in the model, including the "worst case" scenario, assume that no imidacloprid at all passes to waterways from the UK's population of 7.5–12.2 million cats [24, 26].

Unpublished supporting studies providing insufficient evidence

Some of the data used to determine the emission fractions to waterways from pets undergoing the three scenarios are based on unpublished, in-house studies by Bayer that are only superficially described (in Additional file 1) and that bear little resemblance to the real-world scenarios. For example, the 'stroking test' used to derive an estimate of abrasion of imidacloprid from pets to bedding (Anthe et al.'s F_{abr}) involved "stroking the dogs in a standardised manner" four times, but neither the time period nor manner of stroking are stated. Real-world dogs and cats spend many hours in contact with pet bedding, pet clothing, owners' bedding, owners' clothing, hands, furniture and carpets, all of which may be washed. Similarly, the 'immersion test' used to derive an estimate of transfer of imidacloprid from dogs to water during bathing ($F_{washing}$ in Fig. 1) or heavy rain involved immersion of collar-treated dogs in still water for just 5 min—that may result in much less transfer to water than active washing or shampooing of the dog, which may also take longer than 5 min. Additionally, no immersion tests are described for spot-on-treated dogs. Anthe et al. claim that the estimated proportions, derived from these studies and the authors' "expert judgement" are conservative and provide an adequate margin of safety, however this claim is questionable as the true proportions released through these scenarios are unknown, and the authors have a conflict of interest.

Critique of Anthe et al.'s interpretation of their calculated PEC

The above points demonstrate that Anthe et al. made multiple assumptions leading to underestimation by their model of the amount of imidacloprid passing to waterways, and so underestimation of their calculated PEC, and of the contribution of VMPs to the imidacloprid pollution of waterways. Some of these underestimations were made at sequential points in their model, thereby being multiplicative, leading to a potentially large overall underestimation of their calculated PEC. The further criticisms below demonstrate that their discussion and interpretation of the calculated PEC further underestimates the contribution of VMPs to imidacloprid pollution of waterways:

Inappropriate comparison of model PEC to measured pollution levels

Anthe et al. compare the theoretical average PEC of 4.8 ng/l produced by their model to the single highest reported surface water concentration measured in any location in the UK water monitoring programme (190 ng/l), to conclude that the use of imidacloprid as a VMP for companion animals can only explain a very low portion of the measured surface water concentrations. However, their model calculates the emissions from a 'standard' STP, serving a default population of 10,000 people (4000 households), and a default dilution factor of 10 from STP effluent into the adjacent receiving river water is applied to this [23]. By contrast, Somerhill Stream, the sample site at which the highest concentration was measured, is immediately downstream of Tunbridge Wells North STP, which serves a population of 31,441 [13]—and is a small stream [12]. Therefore, the true contribution from VMPs in this location is likely to be far greater than the average PEC their model was designed to estimate, and the comparison—which is included in the abstract of Anthe et al. to support the conclusion that the calculated concentrations were much lower than measured imidacloprid—is inappropriate and misleading.

Furthermore, contrary to Anthe et al.'s statement that it is impossible to 'quantify the extent of emissions' into Somerhill Stream, or to 'identify the major single source of emission', the sources of imidacloprid were investigated in November 2018 when the stream was in a normal flow state. A sample taken in the stream 200 m upstream of the main Tunbridge Wells North Water Treatment Works outflow detected no imidacloprid, a sample taken in the outflow from the sewage treatment works detected 233 ng/l of imidacloprid, and a sample taken downstream of the sewage treatment works at Old Forge Farm Bridge detected 192 ng/l imidacloprid. Two additional inflows were identified in the 1.3 km stretch between the two in-stream sample points, a tributary and an outflow pipe of unknown source—both were tested and no imidacloprid was detected (pers. comm. [29]; Buglife—The Invertebrate Conservation Trust [4]). Therefore, at least on that sampling date, we can be reasonably certain that the imidacloprid emissions were predominantly from the STP. Münze et al. [22] also found substantial contributions to imidacloprid pollution in German streams arose from STPs, and Webb et al. [33] reported an STP as a year-round source of imidacloprid in a stream in Iowa, USA, with their data implicating municipal wastewater effluent as the origin of the imidacloprid. Sadaria et al. [27] found imidacloprid to be ubiquitous in Californian STPs that do not receive outdoor runoff. Their investigation of potential sources suggests that topical pet flea products are likely to be an important household source of imidacloprid transported down-the-drain to STPs, a finding supported by a subsequent study [35].

PNECs

Alongside recognised EU-predicted no effect concentrations (PNECs) for imidacloprid of 4.8 and 8.3 ng/l [5,

11], Anthe et al. include a far higher 'PNEC' of 200 ng/l, based on a Bayer-funded environmental risk assessment [34], derived from mesocosm studies. This latter PNEC appears to be based on the concept of "functional redundancy" in aquatic ecosystems, namely, that the impairment of sensitive species is not expected to alter overall ecosystem function because the ecological functions of those species will be replaced by other functionally similar species. Further, Münze et al. [22] demonstrated that routinely measured neonicotinoid insecticide levels in German streams affected not only aquatic ecosystem composition, but also ecosystem function such as leaf litter breakdown—and that these effects were observed below accepted environmental thresholds. In other words, there is reason to believe that the PNEC produced by the Whitfield-Aslund et al. study significantly exceeds the true NEC (no effect concentration).

Pollution levels associated with STPs

Analysis of the EU watch list water monitoring data presented in Anthe et al. shows that the highest levels of pollution occurred at sites immediately downstream of STPs (Additional file 1: Fig. S1, Table S1, $p < 0.05$, Wilcoxon rank sum test). This is consistent with the findings of Perkins et al. [25] and suggests that STPs are contributing significantly to the pollution. Anthe et al. do not acknowledge or discuss this significant and highly relevant pattern in the data they present. Instead, they use the calculated PEC from their model to argue that VMPs do not contribute substantially to imidacloprid pollution of UK waterways, but do not provide any substantial alternative explanation for the imidacloprid pollution seen, or for why higher levels are consistently found in locations immediately downstream of STPs, other than to conclude that "imidacloprid concentrations in UK surface waters cannot be attributed to a specific end-use of the compound but may result from various applications".

Comparison of model PEC to PNEC

Anthe et al.'s model predicts an environmental concentration of imidacloprid that does not exceed ecotoxicological thresholds. However, their model predicts that bathing dogs alone results in environmental exposure that equals the PNEC for imidacloprid of 4.8 ng/l established by the European Chemicals Agency [11]. Given that several assumptions underlying the model lead to underestimation of the PEC, correction of any of these will result in a PEC that exceeds the PNEC, thereby invalidating Anthe et al.'s conclusion that imidacloprid from flea-control products does not exceed ecotoxicological thresholds in UK waterways.

Conclusion

In summary, we identify several major flaws in the model presented by Anthe et al. that result in underestimation of the contribution of veterinary flea products to waterway pollution. Most notable is the implicit, but incorrect, assumption that imidacloprid applied to pets is only available for release to the environment for 24 h. Adjusting for the deficiencies described above, their model appears consistent with the conclusion that veterinary flea products contribute substantially to imidacloprid waterway pollution in the UK. However, because the model utilises emissions fractions for animals undergoing the different scenarios (for example, bathing) that are extrapolated from unpublished studies bearing little resemblance to the described scenarios, with insufficient evidence provided to support their derivation, we find that the model presented by Anthe et al. provides no reliable conclusions about the contribution of VMPs to the levels of imidacloprid in UK waterways.

Abbreviations

NEC: No effect concentration; PEC: Predicted environmental concentration; PNEC: Predicted no effect concentration; VMP: Veterinary medicinal product.

Supplementary Information

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Additional file 1: Figure S1. Mean sample site concentrations of imidacloprid (2016-2018) presented by Anthe et al (2020, their Table 3). **Table S1**. Mean sample site concentrations of imidacloprid (2016-2018) at various locations in the framework of the European Watch List, presented in Anthe et al (2020, Table 3).

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Authors' contributions

RP and MW wrote the original draft of the commentary, and all authors discussed and contributed to the manuscript. All authors read and approved the final manuscript.

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Availability of data and materials

The datasets used and analysed during the current study are available from the corresponding author on reasonable request.

Declarations

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Competing interests
The authors declare that they have no competing interests.

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