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Commentary
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Fipronil in Surface Water: An Environmental Calamity Remaining Under Radar in the Netherlands

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Commentary

The phenylpyrazole insecticide fipronil is used in Dutch agriculture in seed coating of cabbage, onions and leek, but was also illegally used as treatment against poultry red mite. Crustaceans and aquatic insects have been shown to be highly sensitive to fipronil and its stable degradation products. An environmental quality standard (EQS) for fipronil in Dutch surface water has been set at 0.07 ng/L. As a result of limitations in analytical methodology, with detection limits usually at 10 ng/L or higher, Dutch Water Boards have been unable to demonstrate fipronil in surface water at concentrations up to 150 times above EQS, creating blind spots in most areas of the country.

However, measurable fipronil concentrations of up to 130 ng/L have been recorded in onion cultivation areas in the southwestern province of Zeeland. Similar fipronil concentrations in surface water have been observed in California. Given the cumulative nature of toxicity, the reported fipronil emissions are bound to cause lethal effects in aquatic organisms. Policy implications are discussed.

Fipronil is a potent phenylpyrazole insecticide used in agricultural, urban and domestic environments to control beetles, ticks, fleas, termites and other pests with high efficiency at very low doses. It is used in the Netherlands as a prophylactic seed coating for cabbage, onions and leek [1], but fipronil has also been used illegally in poultry farms as treatment against the red mite [2]. Fipronil operates in insects as an antagonist of the gamma-aminobutyric acid (GABA) receptors [3].

Crustaceans and insects are highly sensitive to fipronil. The IUPAC Pesticide Properties Database of the University of Hertfordshire indicates an oral acute 48 hour LD50 in honey bees *Apis mellifera* of 4 ng/bee and an acute 96 hour LC50 in the aquatic crustacean *Americamysis bahia* of 140 ng/L [4]. Stable degradation products have been shown to be even more toxic than fipronil itself.

Weston and Lydy [5] determined EC50s and LC50s for fipronil and its sulfide and sulfone derivatives for 14 macroinvertebrate species, and the most sensitive species tested, *Chironomus dilutus*, had a mean 96-h EC50 of 32.5 ng/L for fipronil and 7-10 ng/L for its degradates. Moreover, toxicity to insects is time-dependent and follows Haber's Rule [6], indicating cumulative toxicity with exceptionally low thresholds. Cumulative effects of fipronil emissions are therefore a major threat to non-target invertebrates.

In the Netherlands, an environmental quality standard (EQS) for fipronil in surface water was set at 0.07 ng/L [7]. This value was originally derived by applying a safety factor of 2000 to an acute EC50 of 140 ng fipronil/L for the crustacean *Americamysis bahia* (also referred to as *Mysidopsis bahia*). Subsequent studies supported this EQS when a safety factor of 100 was applied to a chronic NOEC of 7.7 ng/L in the same species [2]. However, monitoring fipronil emissions turned out to be a nearly impossible task.

As a result of limitations in analytical methodology, with detection limits usually at 10 ng/L or higher [8], Dutch water boards were unable to demonstrate fipronil in surface water at concentrations up to 150 times above EQS, creating blind spots in most areas of the country [9]. However, measurable fipronil concentrations of up to 130 ng/L have been recorded in onion cultivation areas in the southwestern province of Zeeland [8] (Figure 1 & Table 1).

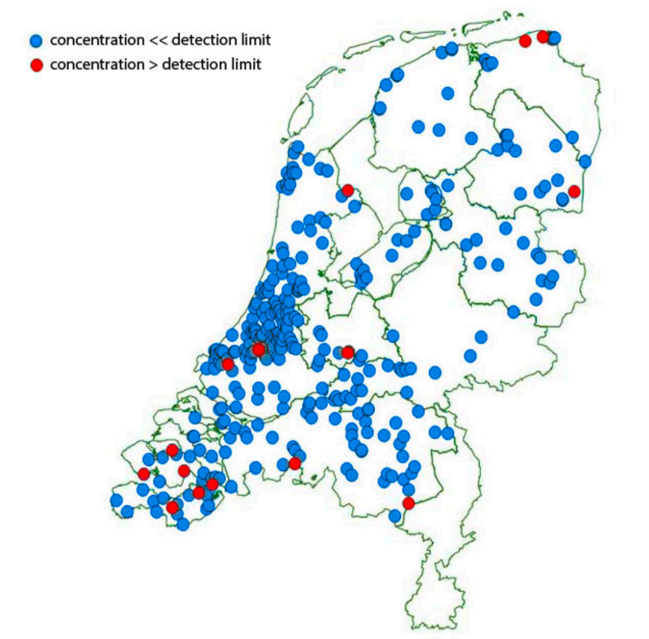

Figure 1: Fipronil determinations in Dutch surface water in 2016 [8]. The detection limit was 10 ng/L or higher. The EQS is 0.07 ng/L.

Similar fipronil emissions have been demonstrated in California. In 2003, the California Department of Pesticide Regulation (DPR) began to allow professional applicators to spray fipronil around buildings to control nuisance insects [5]. Fipronil was measurable in 88% of the samples collected from sixteen urban waterbodies during or immediately after rain events, with a maximum concentration of 49.1 ng/L, and a median concentration of 21.2 ng/L [5]. In samples from Bay Area storm drains and creeks in two watersheds collected between 2008 and 2011, Ensminger et al. measured fipronil concentrations up

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to 460 ng/L [10]. In an intensive two-year sampling program in Sacramento and Orange Counties [11], median concentrations of fipronil plus its three degradation products in runoff were 14 to 441 ng/L.

Sampling location code	Date of sampling	Fipronil concentration (ng/L)	96-h EC50 in Chironomus dilutes (ng/L) **		
			Fipronil	Fipronil-sulfon	Fipronil-sulfide
MPN 10236	7-Apr-16	17	30-35	7.5-7.9	9.3-10.5
MPN 10236	unidentified	28			
MPN 5650	7-Jun-16	16			
MPN 5650	unidentified	20			
MPN 8130	1-Aug-16	19			
MPN 8130	unidentified	14			
MPN 9119	10-Jun-16	130			
MPN 1442	7-Oct-16	11			
*source: Waterboard Scheldestromen [8] **source: Weston & Lydy [5]					

Table 1: Fipronil concentrations in surface water of onion cultivation areas in Zeeland.

Given the cumulative nature of toxicity [6], the reported fipronil emissions are bound to cause lethal effects in aquatic organisms. A conservative EQS for a pesticide, however prudent as such, is self-defeating in practical terms when adherence cannot be effectively monitored, and may lead to ecotoxicity, as in the case of fipronil. The evidence shows that registration of fipronil as a seed coating for onion cultivation is associated with unacceptable risk and cannot be justified. Furthermore, to obtain market authorization, pesticide producers should be obliged to develop analytical methodology for effective monitoring of emissions. Li and Jennings [12] demonstrated that there is a large variance of pesticide regulatory standard values promulgated by global regulatory agencies including the Netherlands National Institute for Public Health and the Environment. There is an urgent need for harmonization in this respect to set clear goals for pesticide development.

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