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REVIEW

The Fate of Fluopyram in the Soil–Water–Plant Ecosystem: A Review

Paresh H. Rathod[1,2] · Paresh G. Shah[2] · Kaushik D. Parmar[2] · Ravi L. Kalasariya[2]

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Abstract
This review aims to discuss all likely pathways of the environmental fate of fluopyram to enable a better understanding of the probable ecological risks associated with its agricultural usage. The fluopyram is a broad-spectrum molecule to control various fungal plant pathogens as well as nematodes. It belongs to a new chemical class named 'pyridinyl ethylbenzamides'. The literature review has shown that the sorption–desorption, degradation and leaching of fluopyram differed among the soil types, and much is still to be studied concerning the fate of fluopyram in different types of the soil environment. Indeed, research suggests that the high persistent behaviour of fluopyram particularly in soil and water/sediment environment can present environmental risks. Hence, with a foreseen widespread and substantial use of fluopyram, it would be indeed crucial to assess the possible environmental risks due to injudicious usage of fluopyram.

Abbreviations

ai	Active ingredient
APVMA	Australian Pesticides and Veterinary Medicines Authority Declaration
ATP	Adenosine triphosphate
BZM	Benzamide
CIB&RC	Central Insecticide Board and Registration Committee, India
DAR	Draft Assessment Report, European Union
DFOP	Double first-order in parallel
DT_{50}	Half-life, Time required for 50% degradation of the initial amount
DT_{75}	Time required for 75% degradation of the initial amount
DT_{90}	Time required for 90% degradation of the initial amount
ECHA	European Chemicals Agency
EFSA	European Food Safety Authority
EPA	Environmental Protection Agency, USA
FAO	Food and Agriculture Organization
IUPAC	International Union of Pure and Applied Chemistry
JMPR	The Joint FAO/WHO Meeting on Pesticide Residues
K_d	Coefficient of distribution
K_{OC}	Organic carbon normalized coefficient of distribution
LD	Lethal dose
LOQ	Limit of quantification
MRL	Maximum residue limit
NYSDEC	New York State Department of Environmental Conservation
NZ-EPA	New Zealand Environmental Protection Authority
PAA	Pyridylacetic acid (a metabolite of fluopyram)
PCA	Pyridyl carboxylic acid (a metabolite of fluopyram)
PHI	Pre-harvest interval
PMRA	Pest Management Regulatory Agency, Canada
SC	Suspension concentrate
SDH	Succinate dehydrogenase
SDHI	Succinate dehydrogenase inhibitor
SFO	Single first-order
TRR	Total radioactive residues
VKM	The Norwegian Scientific Committee for Food & Environment
WHO	World Health Organization

✉ Paresh H. Rathod
rathod23904@aau.in

Paresh G. Shah
paresh243@aau.in

Kaushik D. Parmar
drkdp28@gmail.com

Ravi L. Kalasariya
dr.ravi@aau.in

[1] Main Forage Research Station, Anand Agricultural University, Anand, Gujarat 388 110, India

[2] AINP on Pesticide Residues, Anand Agricultural University, Anand, Gujarat 388 110, India

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Introduction

In recent years, several new fungicides that inhibit the succinate dehydrogenase (SDH) enzyme, collectively termed as succinate dehydrogenase inhibitor (SDHIs), are the fastest growing in terms of new compounds being developed and launched into the agriculture market (Sierotzki and Scalliet 2013; Xiong et al. 2015). Fluopyram, being the SDHI compound and the first of a new group of fungicide—called pyridinyl ethylbenzamides, have been quickly adopted by the agriculture market. The molecule was discovered and developed by Bayer CropScience in 2007 (Fought et al. 2009) for controlling several plant disease-causing pathogens (such as *Botrytis cinerea*, powdery mildew, *Sclerotinia* spp. and *Monilinia* spp.) of horticultural, field and vegetable crops (Veloukas and Karaoglanidis 2012; Jeschke 2016). To this, Kandel et al. (2018) have reviewed over 200 studies about the effects of fluopyram-amended seed treatment on sudden death syndrome (caused by a soilborne fungus, *Fusarium virguliforme*) and yield of soybean, and results of a meta-analysis of these data showed that the seed treatment with fluopyram reduced the foliar disease index by 35% and increased the soybean yield by 295 kg ha^{-1} (i.e. by 7.6%) in comparison to standard seed treatments. Fluopyram is highly effective even at low application rates, both on its own and in co-formulations with other fungicides such as tebuconazole, prothioconazole, pyrimethanil and trifloxystrobin (Labourdette et al. 2010).

Subsequently, the potential of fluopyram as a nematicide was discovered and it was found effective against root-knot nematodes (Faske and Hurd 2015; Jones et al. 2017; Beeman et al. 2019; Ji et al. 2019; Watson and Desaeger 2019). Bayer CropScience recently has launched its new nematicide based on the active ingredient fluopyram, marketed under the brand name "Velum", is the first nematicide acting via complex-II inhibition (Jeschke 2016; Oka 2020). Owing to fungicidal as well as nematicidal actions, and further, it can be used as either foliar, seed treatment or in-furrow soil application, there has been a growing interest in fluopyram for vegetable and fruit crops, and henceforth fluopyram market is likely to boost over next few years. In India, fluopyram 200 g L^{-1} SC (i.e. 17.7% SC) has recently been registered as a fungicide as co-formulation with tebuconazole for controlling post-harvest diseases such as black mould and neck rot in onion (CIB&RC 2018a); false smut and dirty panicle in rice (CIB&RC 2018b); powdery mildew and anthracnose in chilli and grape (CIB&RC 2017); and fluopyram 34.48% SC as a nematicide for the use against root-knot nematodes in tomato crop (CIB&RC 2018c).

Although most plant protection chemicals are intended to break down over time to prevent the accumulation of their residues in a soil–water–plant ecosystem, it has been found that several chemicals persist longer than expected and could pose potential threats to human beings (Hazlett 2003). Looking at the environmental risks and food safety issues associated with the injudicious use of such persistent molecules, it is essential to review the fate of agrochemicals in soil, water and plant for their effective usage and as well for environmental safety.

Fluopyram has been classified as environmentally safe due to its relatively low median lethal dose (LD_{50}) for mammals (for rats > 2000 mg kg^{-1} body weight), however, few studies/reports have questioned the efficacy of fluopyram due to its possible leaching, toxicity and thereby associated human risks (EPA 2012; Rouquie et al. 2014; Tinwell et al. 2014; Bénit et al. 2018). As per the reports of Health Canada PMRA (2014, 2016), fluopyram presents a negligible risk to soil organisms, bees, beneficial arthropods, fish, invertebrates, algae and aquatic plants. Interestingly, soil treated with fluopyram (0–5 mg kg^{-1}) could stimulate the pepper plant growth as well as it could also increase relative abundances of P-solubilizing and N-fixing microbes (Sun et al. 2020). In contrast, Zhang et al. (2014) and Li et al. (2020a,b) have found a harmful effect of fluopyram on overall soil microbes, and it can alter the diversity of the soil microbial community. Fluopyram was found to be slight to highly persistent in different types of soil at European and US field sites with half-lives (DT_{50}) ranging from 21 to 386 and 24–539 days, respectively (JMPR 2010a; DAR 2011a; APVMA 2015). Whereas, the DT_{75} values of fluopyram dissipation in US soils ranged from 521 to > 1000 and the DT_{90} values in the European field soils ranged from 487 to > 1000 days (JMPR 2010a; DAR 2011a), indicating a high potential for fluopyram residues to carry over to the succeeding crops and produces. Further, it is moderately mobile within soils and in certain situations, there could be a possible risk of leaching of fluopyram into deeper soil layers, where it could end up in groundwater (EPA 2012; Health Canada PMRA 2014, 2016; VKM 2014). However, fluopyram is stable to hydrolysis, photolysis, aerobic/anaerobic biotransformation in soils and it is also persistent in aquatic systems (Health Canada PMRA 2014, 2016; Sjerps et al. 2019).

Being a new molecule, scientific literature on the dynamics of fluopyram in the environment is limited. Therefore, it is worthy to review the fate (e.g. sorption–desorption, dissipation, leachability, etc.) of fluopyram in the soil–water–plant system.

Fluopyram—Properties and End-Use Products

Fluopyram, a common name for *N*-{2-[3-chloro-5-(trifluoromethyl)-2-pyridyl]ethyl}-α,α,α-trifluoro-*o*-toluamide (IUPAC name), is a novel broad-spectrum systemic compound that

belongs to new chemical group pyridinyl-ethyl-benzamide (or pyramides) within the SDHIs class. It is the only member of the pyramides group and differs in shape and molecular flexibility from other SDHI fungicides. It is usually used as a fungicide against fungal plant diseases such as *Botrytis*, powdery mildew, apple scab, *Alternaria*, *Sclerotinia* and *Monilinia* and also as a nematicide against root-knot nematodes. A detailed physical and chemical characteristics of the fluopyram are given in Table 1 (JMPR 2010a; DAR 2011b).

Products containing fluopyram are currently registered worldwide including in Canada, the USA, the European Union (EU), Australia, New Zealand, China and India (Table 2). Fluopyram is registered in the various co-formulated products with other fungicides and/or insecticides such as tebuconazole, prothioconazole, pyrimethanil, trifloxystrobin, triadimenol and imidacloprid. Fluopyram is generally formulated as 400–500 g ai L^{-1} (w/v) suspension concentrate (SC), containing 34.48–41.70% (w/w) fluopyram. While, combination products with 1:1 pre-mix of fluopyram and prothioconazole, tebuconazole or trifloxystrobin and a 1:3 pre-mix of fluopyram and pyrimethanil are also commonly formulated. An active ingredient fluopyram is still patent-protected in Germany, France, the United Kingdom, Canada and other countries until at least 2023, while it is patent-protected until 2024 in the USA and 2025 in Brazil (Bayer 2020). Several fluopyram-containing end-use products are currently registered around the world, the majority of which are produced by Bayer CropScience Ltd., BASF, and FMC Corporation (Table 2).

Fluopyram—Mode of Action

Fluopyram, as a third-generation SDHI compound, has broad-spectrum activity against different fungal species for a variety of crops (Beckerman 2013). The SDHI fungicides target the enzyme succinate dehydrogenase (SDH enzyme, also termed as complex-II or succinate-ubiquinone oxidoreductase; Fig. 1), which is found in the inner mitochondrial membrane of eukaryotes. It is the only enzyme that acts as a ubiquitous and key element for both tricarboxylic (or Krebs) cycle and mitochondrial electron transport chain of fungal pathogens (Kuhn 1984; Avenot and Michailides 2010). Enzyme SDH consists of four subunits (A, B, C and D), and fluopyram targets and binds to ubiquinone binding site, which is formed by the subunits B, C and D of complex-II and thereby inhibits respiration (i.e. succinate to fumarate oxidation) of phytopathogenic fungi by blocking electron transport (Fig. 1). Fluopyram moves systematically within the plants through a multidimensional penetration as well as an acropetal redistribution (Dubournet et al. 2012). Fluopyram acts against nematodes in the same way as it does against fungi, by inhibiting cellular respiration and cellular respiration (Burns et al. 2015; Heiken 2017; Hawk 2019; Oka 2020).

Fluopyram—Toxicological Data and Residue Definition

As per the reports of the Joint FAO/WHO meeting on pesticide residues (JMPR 2010b,c), fluopyram had low acute toxicity by the oral, dermal and inhalational routes in rats. Further, it was not a skin or eye irritant in rabbits and was

Table 1 Physical and chemical characteristic of 99.8% pure fluopyram

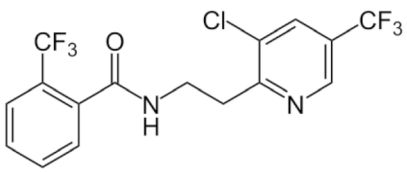
Molecular structure and formula	 $C_{16}H_{11}ClF_6N_2O$ $C_{16}H_{11}ClF_6N_2O$
Molecular mass	396.72 g mol^{-1}
Physical state (appearance)	White powder with no noticeable odour
Solubility in water	15–16 mg L^{-1} at 20 °C (pH 4–9)
Solubility in organic solvents	> 250 g L^{-1} at 20 °C in Methanol, Acetone, Ethyl acetate, Dichloromethane, Dimethyl Sulfoxide
Melting point	117.5 °C
Boiling point	318–321 °C
Dissociation in water	No pKa at environmentally relevant pH
Vapour pressure	1.2×10^{-6} Pa at 20 °C; 3.1×10^{-6} Pa at 25 °C (low volatility)
Henry's law Constant	2.98×10^{-5} Pa m^3 mol^{-1} at 25 °C (non-volatile)

Table 2 Worldwide registered end-use products containing fluopyram

Registered product name	Technical ingredient(s) and concentration	Registered for use on crops…	Country
The product containing Fluopyram alone			
ILeVO®	Fluopyram 48.4% SC	Soybean	USA
ILEVO® Seed Treatment	Fluopyram 49.02% SC	Soybean	USA
COPeO® Prime	Fluopyram 48.4% SC	Cotton seed	USA
CoPeO® Seed Treatment	Fluopyram 49.02% SC	Cotton seed	USA
Indemnify®	Fluopyram 34.50% SC	Turfgrass	USA
Luna® Privilege	Fluopyram 41.50% SC	Apple, Pears, Cherry, Dried beans, Peanut, Potato, Strawberry, Sugarbeet, Tree nut, Watermelon, Wine grape	USA, UK
		Banana, Kiwifruit, Greenhouse fruiting vegetables (except cucurbits)	Australia, New Zealand
Velum® Velum® One	Fluopyram 41.50% SC	Fruit, field and vegetable crops	USA
Velum® Prime	Fluopyram 41.50% SC	Brassica leafy vegetables, Bulb vegetables, Caneberries, Citrus, Cucurbits, Fruiting Vegetables, Potato, Stone fruits, Sweet potato, Tobacco, Pome Fruit, Strawberry	USA, Canada
	Fluopyram 34.48% SC	Potato, Carrot	UK
		Tomato	India
		Chili, Potato, Melon, Tomato, Carrot	Indonesia
		Carrot (except baby carrot)	New Zealand
Verango® Prime	Fluopyram 41.50% SC	Potato, Coffee, Sugarcane, Soybean	Brazil
The product containing Fluopyram with other Fungicide or Insecticide ingredients			
Broadform®	Fluopyram 21.40% + Trifloxystrobin 21.40% SC	Ornamentals and crops in residential and landscapes crops grown in protected structures	USA
Delaro® Complete	Prothioconazole 14.90% + Trifloxystrobin 13.10% + Fluopyram 10.90% SC	Corn, Soybean, Sweet corn, Wheat	USA
Exteris® Stressgard®	Fluopyram 1.19% + Trifloxystrobin 1.92% SC	Turfgrass	USA
Ascra® Xpro Macfatre® Xpro	Bixafen 6.4% + Fluopyram 6.4% + Prothioconazole 12.7% SE (Suspo-emulsion)	Wheat, Barley, Oats	UK, Ireland
Raxil® Star (seed treatment)	Fluopyram 1.72% + Prothioconazole 8.62% + Tebuconazole 5.17% SC	Barley, Oats, Ryecorn, Triticale, Wheat	UK, Ireland, New Zealand
Luna® Sensation	Fluopyram 21.4% + Trifloxystrobin 21.4% SC	Almond, Artichoke, Low growing berry, Brassica—head, stem and leafy greens, Carrot, Cherry, Dill seed, Citrus, Pome, Small vines, Herbs, Hops, Leafy greens, Leaf petioles, Melons, Nut trees, Pistachio, Tomato, Root vegetables	USA, Canada
		Almonds, various fruit crops and lettuce, Grapes, Stone fruit	Australia, New Zealand
	Fluopyram 25% + Trifloxystrobin 25% SC	Strawberry, Pepper and Chilli	UK
Luna Smart™	Fluopyram 21.4% + Trifloxystrobin 21.4% SC	Apple, Shallot, Chili and Mango	Indonesia

Table 2 (continued)

Registered product name	Technical ingredient(s) and concentration	Registered for use on crops…	Country
Luna® Experience	Fluopyram 17.7% + Tebuconazole 17.7% SC	Grapes, Chilli, Onion, Paddy	India
		Almond, Brassica leafy beans, Bulb vegetables, Cucurbits, Grape, Okra, Sunflower, Stone fruits, Tree nuts, Pistachio	USA
		Banana	Australia
Kivalo®[a]	Fluopyram 17.7% + Tebuconazole 17.7% SC	Grapes, Chilli, Onion, Rice	India
Luna® Tranquility	Fluopyram 11.3% + Pyrimethanil 33.8% SC	Low growing berry, Bushberry, Pome, Ginseng, Lemons, Onion—bulb and green, Tomato, Root vegetables, Tuberous & corm vegetables	USA, Canada
Luna® Devotion	Fluopyram 21.6% + Triadimenol 21.6% SC	Onion	New Zealand
Proline® GOLD Propulse®	Fluopyram 17.4% + Prothioconazole 17.4% SC	Bush berries, Canola, Corn, Cotton, Cucurbits, Dried beans, Peanut, Rapeseed, Soybean, Sugarbeet	USA, Canada
	Fluopyram 12% + Prothioconazole 12% SE	Oilseed rape	UK
Velum® Total	Fluopyram 15.4% + Imidacloprid 22.2% SC	Cotton, Peanut	USA

Source Product label or SDS, Bayer CropScience Webpages of respective countries

[a]Manufactured by FMC Corporation India

not a skin sensitizer in mice. Similarly, various environmental, food safety and/or health agencies of Australia (APVMA 2015), Canada (Health Canada PMRA 2014, 2016), European Union (DAR 2011c; ECHA 2013; EFSA 2013), New Zealand (VKM 2014; NZ-EPA 2015a,b, 2018a,b) and USA (NYSDEC 2017; EPA 2019) reported the toxicological assessments of fluopyram and/or its formulated products. Table 3 summarizes the toxicological details of fluopyram.

The parent compound 'fluopyram' was found as a major residue in all of the plant commodities studied (primary and rotational crops, food and feed), and it was detected at a significant level in all trials (JMPR 2010a; DAR 2011d). In various supervised crop field trials, residues of fluopyram-benzamide (BZM), pyridyl carboxylic acid (PCA) and to a lesser extent pyridylacetic acid (PAA) were also detected in many commodities, often at longer pre-harvest intervals (PHIs) of 10–21 days and levels of > 0.02 mg kg^{-1} (JMPR 2010a). Higher amounts of BZM and less frequently, PCA and its methyl sulfoxide (0.1 mg kg^{-1}) and PAA (rarely > 0.05 mg kg^{-1}) were found in some legumes and brassicas, rapeseed, grapes, lettuce and strawberries. The parent residue usually detected in greater quantities than its metabolites (JMPR 2010a; DAR 2011d). According to the report of JMPR (2010a), the residue definition for maximum residue limit (MRL) compliance for plants products should be 'fluopyram'. Similarly, in the evaluation report of Health Canada PMRA (2014), a residue definition for enforcement/monitoring purpose is 'fluopyram' for plant commodities, whereas for risk assessment is 'fluopyram plus metabolite BZM' for legume vegetables, oilseeds crops and 'fluopyram' in all other plant commodities. The European Food Safety Authorities have also proposed similar definitions. (DAR 2011d; EFSA 2013).

Moreover, two of the main metabolites in plants (i.e. BZM and PCA) were also observed in animal metabolism studies (JMPR 2010a). But, it has been confirmed that the metabolite PCA and its methyl sulfoxide are significantly less toxic than its parent fluopyram (JMPR 2010b; APVMA 2015). Therefore, the BZM, PCA and PAA metabolites are not considered in residue definitions for plant commodity for MRL enforcement or estimation of dietary intake. Table 3 summarizes the residue definitions for various commodities and entities as deliberated in these reports.

Fluopyram—Environmental Fate

While in the ecosystem, the pesticides dissipate by a variety of mechanisms viz., degradation, sorption–desorption, volatilization, plant uptake and/or metabolism, runoff to surface waters and leaching to groundwater. Among these mechanisms, pesticide degradation is a key process that determines the environmental fate and transport of

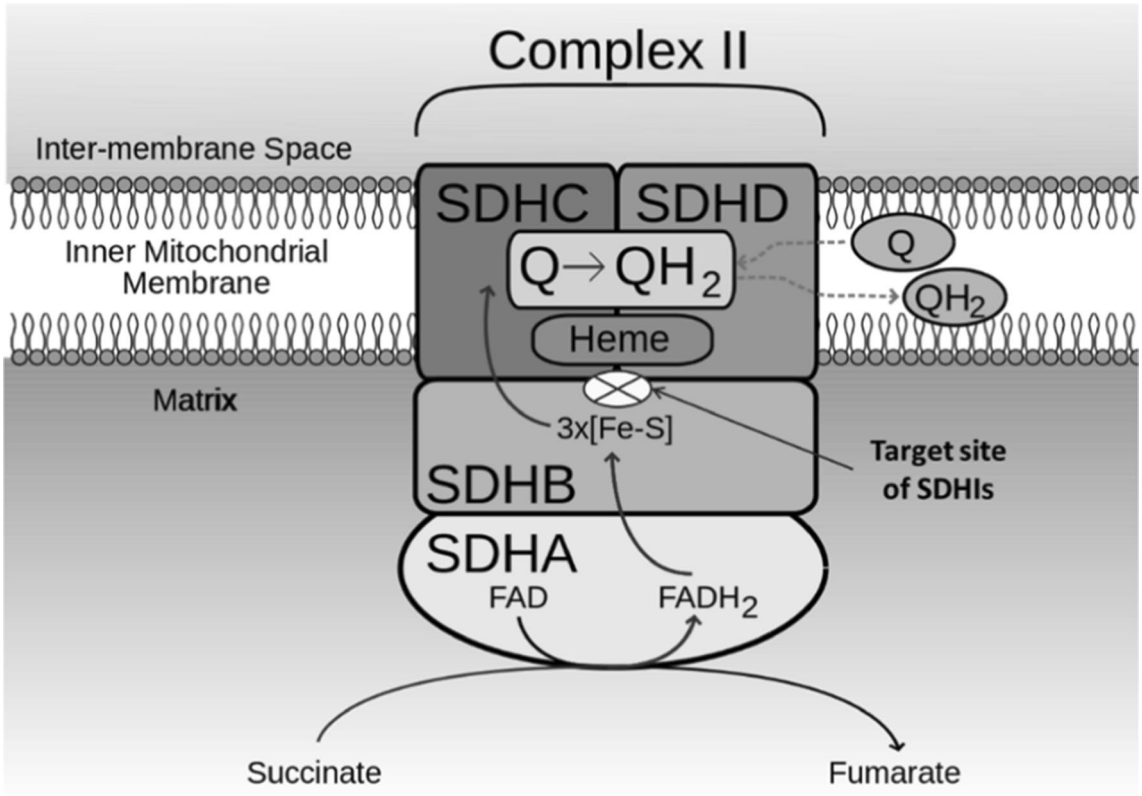


Fig. 1 The succinate dehydrogenase (complex-II) and the target site of fluopyram (Source: Johnhfst, http://en.wikipedia.org)

pesticide, and it includes various processes such as abiotic degradation (e.g. oxidation, hydrolysis and photolysis) and biodegradation. Through these processes, a pesticide molecule is either transformed into a degradation product or completely mineralized to carbon dioxide. The faster a pesticide degrades, the less time it stays in the applied field for the pesticide activity or another movement. Furthermore, the environmental fate of pesticides is determined by the chemical properties of molecules (e.g. ability to bind to soil, susceptibility to degradation), its formulation, dose and mode of application, and environmental factors (e.g. soil types, rainfall, topography, agricultural management practices).

Based on the physical and chemical characteristics of fluopyram (Table 1) and according to the existing reports (JMPR 2010a; DAR 2011a; Health Canada PMRA 2014; APVMA 2015), fluopyram is moderately soluble in water (~ 15 mg L^{-1} at 20 °C), unlikely to volatilize from moist soil (1.2×10^{-6} Pa at 20 °C) or water surfaces (non-dimensional Henry's Law Constant H = ~ 1.2×10^{-8} at 20 °C). Fluopyram has a limited potential for photo-transformation in the environment as it does not dissociate under environmentally relevant pH conditions. Based on the n-octanol/water partition coefficient (log K_{OW} = 3.3), fluopyram has the potential to bioaccumulate and is expected to have low soil mobility.

Since fluopyram is intended for use as a foliar treatment, seed treatment, drip irrigation, or soil drenching, it is important to consider the different pathways of environmental fate, such as hydrolysis, aerobic soil degradation, soil dissipation, soil photolysis, adsorption/desorption in soils, leaching within the soil profile and into groundwater, and degradation in filed crops (i.e. plant metabolism). The environmental fate of fluopyram has been investigated in systematic series of laboratory and field studies (the majority of which were performed with ^{14}C-labelled active substance: phenyl-UL-^{14}C-fluopyram pyridyl-2,6-^{14}C-fluopyram) and have been briefly discussed in the evaluation reports of the respective environmental or health authorities of Australia, European Union, Canada, the USA, Germany and others (JMPR 2010a; DAR 2011a; Health Canada PMRA 2014; APVMA 2015).

Dissipation of Fluopyram in Soils

Abiotic Transformation: Hydrolysis and Photolysis

Fluopyram is hydrolytically stable under acidic, neutral and alkaline conditions since no major transformation products were detected at all relevant pH conditions,

Table 3 Summary of toxicological data of fluopyram

Absorption, distribution, excretion and metabolism in mammals	
Rate and extent of oral absorption	Rapid; absorption approximately 93%
Distribution	Wide; highest concentration in liver, kidney
Rate and extent of excretion	> 95% within 168 h (35–60% in urine; 39–64% in faeces; up to 79–7% in bile)
Metabolism in animals	Extensive; hydroxylation; oxidation and hydrolytic cleavage of the molecule, followed by conjugation (glucuronic acid, sulphate)
Toxicological major compounds (animals, plants and environment)	Fluopyram
Acute toxicity	
Rat, Lethal dose LD_{50}, oral	> 2000 mg kg^{-1} body weight (bw)
Rat, LD_{50}, dermal	> 2000 mg kg^{-1} bw
Rat, Lethal concentration LC_{50}, oral	> 3.4–5.1 mg L^{-1} (4 h, nose-only exposure)
Rabbit, dermal irritation	Not irritating
Rabbit, ocular irritation	Minimally irritating
Mouse, dermal sensitization	Not sensitizing
Short-term studies of toxicity	
Rat, Lowest oral no-observed-adverse-effect level (NOAEL)	12.5 mg kg^{-1} bw day^{-1} (90-day study)
Rat, Lowest dermal NOAEL	300 mg kg^{-1} bw day^{-1} (28-day study)
Long-term toxicity, carcinogenicity and genotoxicity	
Rat, Lowest NOAEL	1.2 mg kg^{-1} bw day^{-1} (2-year study)
Carcinogenicity	Unlikely to pose a carcinogenic risk to humans at levels of dietary exposure
Other toxicological studies	
Studies on plant metabolites	PCA and fluopyram-methyl sulfoxide; both have lower toxicity than the parent compound and not genotoxic in vitro
Reference values	
Acceptable daily intake (ADI)	0.012 mg kg^{-1} bw day^{-1}
Acceptable operator exposure level (AOEL)	0.03–0.125 mg kg^{-1} bw day^{-1}
Acute reference dose (ARfD)	0.25–0.50 mg kg^{-1} bw day^{-1}
Residue definitions for monitoring purposes	
Food of plant origin	Fluopyram
Food of animal origin	Sum of fluopyram and metabolite BZM, as fluopyram
Soil	Fluopyram
Water (surface and ground/drinking)	Fluopyram
Residue definitions for risk assessment	
Food of plant origin	Sum of fluopyram and metabolite BZM, as fluopyram
Food of animal origin	Sum of fluopyram, metabolites BZM and fluopyram-E/Z-olefine isomers, as fluopyram

hence, no half-life could be calculated. The reports of JMPR (2010a), DAR (2011a) and APVMA (2015) reviewed several studies of photolysis of phenyl-UL-^{14}C-fluopyram in sandy loam soil at 20 °C, and at 75% soil moisture. The result shows that the fluopyram was stable to photolysis, and no soil degradation of radio-labelled fluopyram applied at 7.8 µg g^{-1} (an equivalent to 0.25 kg ai ha^{-1}) was observed even after artificial irradiation (< 290 nm with light intensity 276 W m^{-2}) for 23 days. Thus, hydrolysis and photolysis of fluopyram in/on the soil surface are not important degradation pathways in the terrestrial environment.

Biotransformation: Degradation Pathways in Soil

Degradation of fluopyram in soil mainly occurs through aerobic soil metabolism that involves hydroxylation in the 7-position of fluopyram to form hydroxylated metabolite, fluopyram-7-hydroxy (identified in all tested soils with maximum 4.2% of phenyl label to 3.3% of applied pyridyl label), and which in turn is cleaved to form, respectively, the metabolites BZM and PCA (which further degrades to methyl sulfoxide) followed by microbial mineralization to CO_2 and formation of non-extractable residues (JMPR 2010a; DAR 2011a). Figure 2 shows degradation routes of fluopyram, and generally, four minor metabolites that

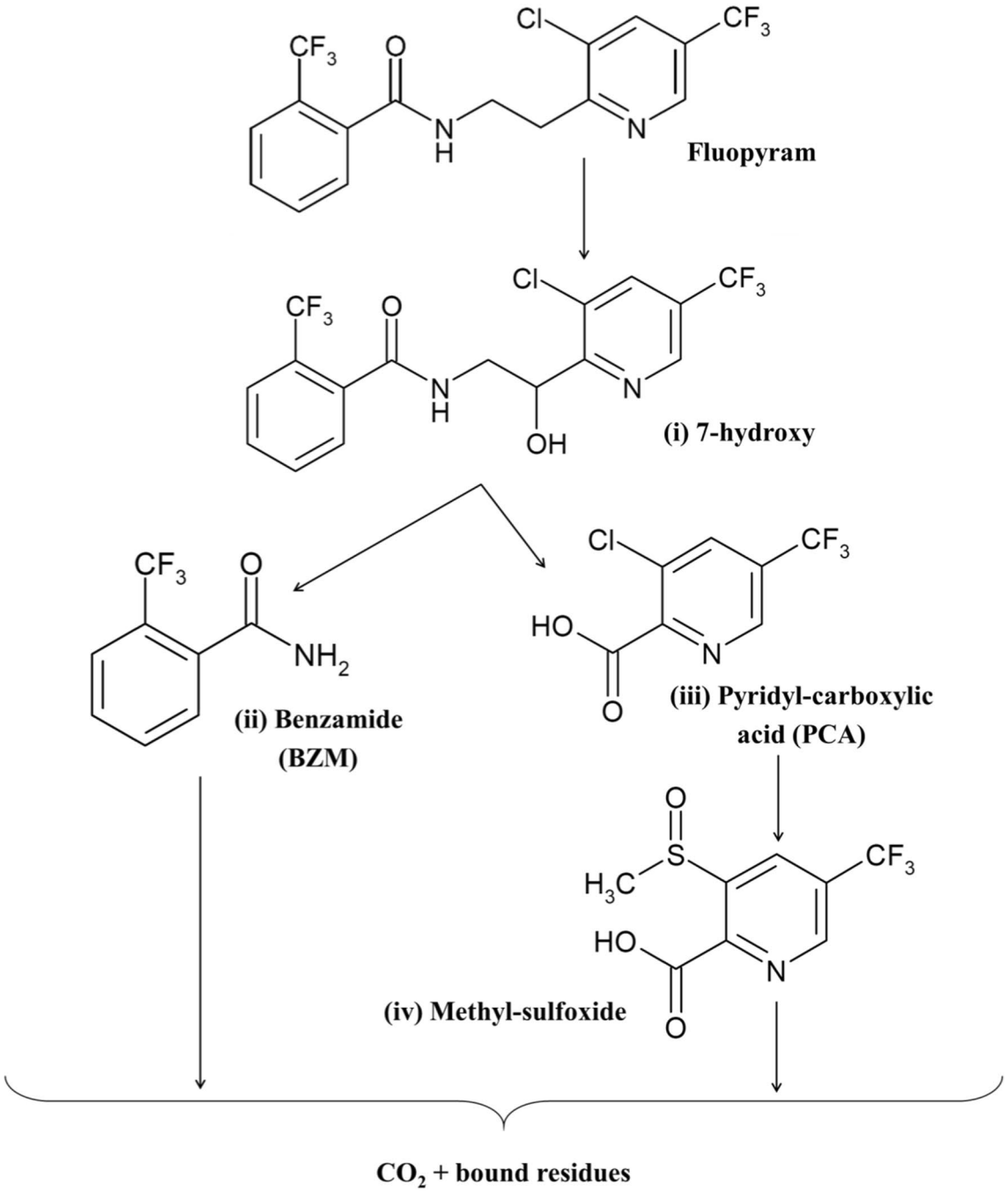


Fig. 2 Degradation pathway of fluopyram in soils (JMPR 2010a; DAR 2011a)

expected to form in soils are (i) fluopuram-7-hydroxy, (ii) fluopyram-BZM, (iii) fluopyram-PCA and (iv) methyl sulfoxide. Fluopyram in different types of soil was found to be hydrolysed to 7-hydroxy by 62 days (maximum of 4.2% AR, applied radio-labelled fluopyram); to benzamide (maximum 1.1% AR) by 30–121 days; to pyridyl carboxylic acid (PCA; maximum 0.7% AR) by 30 days; and to the methyl sulfoxide (maximum 1% AR) by 128 days (JMPR 2010a; DAR 2011a).

Rate of Degradation in Soils—Laboratory Studies

The fluopyram was found to be stable, with no significant degradation observed in anaerobic (flooded) German silt

loam soils; and < 1.1%/0.8% (phenyl/pyridyl label) CO_2, and no volatile organic compounds produced throughout the anaerobic conditions (DAR 2011a; APVMA 2015). The single first-order (SFO) half-life of each labelled fluopyram was estimated > 1000 days (determined by extrapolation) and thus fluopyram is considered as stable under anaerobic conditions in the tested soil (DAR 2011a; APVMA 2015).

The results of laboratory aerobic degradation of labelled fluopyram in four German soils and two US soils have been discussed in various evaluation reports (JMPR 2010a; DAR 2011a; EFSA 2013; APVMA 2015) and based on the best-fitted SFO and double first-order parallel (DFOP) kinetic models, values for dissipation time (DT_{50} and DT_{90}) for fluopyram are summarized in Table 4.

The result shows that the fluopyram degraded slowly in soils under aerobic conditions with DT_{50} values ranging from 162 to 464 days in German soils (with mean DT_{50} of 271 days for pyridyl label and 239 days for phenyl label), and 561 to 746 days (with mean DT_{50} of 572 days for pyridyl label and 700 days for phenyl label) in US soils. According to the persistence classification of FAO (2000a,b), greater half-lives indicate that the fluopyram was persistent in most examined soils (DT_{50} > 180 days), but it was found moderately persistent (DT_{50}: 45–180 days) in German clay loam and loam soils. The greater DT_{90} values of 538 to > 1000 days indicated that the fluopyram residues might be carried over to the succeeding crops. In another year-long in vitro study with two US soils as discussed in the report of JMPR (2010a), authors have estimated the SFO-based fluopyram degradation half-lives of 922 days (California sandy loam soil) and 484 days (Nebraska silty clay loam soil).

Some controlled pot or incubation studies showed that the rate of fluopyram degradation was dependent on its initial concentration, and demonstrated a relatively lesser to moderate persistent nature of fluopyram in the studied soils (Zhang et al. 2014; Sun et al. 2020). In the incubation study using silt loam soil (pH 7.8, organic carbon, OC 1.77%) with the spiking of fluopyram at three rates of 0.5, 1.5 and 5 mg kg^{-1}, Zhang et al. (2014) have observed the first-order degradation of fluopyram in soil with half-lives of 64.2, 81.5 and 93.6 days, respectively, and by the 90th day, the corresponding fluopyram concentration in soil was 0.186, 0.620 and 2.481 mg kg^{-1}. The higher concentration of fluopyram (i.e. 5 mg kg^{-1}) possibly retrained the degradation activity of soil microorganisms, and thus authors had observed a relatively longer half-life (Zhang et al. 2014). However, in another pot study using silt loam soil (pH 6.8, OC 2.53%) and pepper as test crop with a similar set of fluopyram applications, Sun et al. (2020) had observed fairly shorter half-lives of 4.2, 7.4 and11.2 days, respectively, for fluopyram application at 0.5, 1.5 and 5.0 mg kg^{-1}. By 45 days of the study period, the fluopyram residues in soil were not detected for 0.5 and 1.5 mg kg^{-1} application rate while 0.26 mg kg^{-1} of residue was detected for 5 mg kg^{-1}. The author had explained such shorter half-lives of fluopyram by the fact of (i) relatively higher temperature, humidity and good light conditions

Table 4 The DT_{50} and DT_{90} values for fluopyram in aerobic soils under laboratory conditions

Country; soil type; soil property	DT_{50} (day)	DT_{90} (day)	Model used
Soil aerobic: pyridyl-2,6-^{14}C-fluopyram			
German; silt loam; pH 6.7, OC 2.4%	210	697	SFO[a]
German; sandy loam; pH 6.2, OC 2.2%	464	> 1000	SFO
German; clay loam; pH 5.2, OC 1.8%	250	829	SFO
German; silty clay loam; pH 7.3, OC 5.1%	162	538	SFO
US; sandy loam; pH 7.9, OC 0.5%	561	> 1000	SFO
US: silty clay loam; pH 6.5, OC 1.7%	583	> 1000	DFOP[b]
Soil aerobic: phenyl-UL-^{14}C-fluopyram			
German; silt loam; pH 6.6, OC 2.1%	221	735	SFO
German; sandy loam; pH 6.6, OC 1.5%	231	769	SFO
German: loam; pH 5.5, OC 2.0%	339	> 1000	SFO
German; loam; pH 6.6, OC 1.3%	165	549	SFO
US; sandy loam; pH 7.9, OC 0.5%	746	> 1000	SFO
US: silty clay loam; pH 6.5, OC 1.7%	654	> 1000	DFOP

[a]SFO (single first-order) kinetic model is a simple exponential equation with two parameters (k and C_0), so-called a simple or single first-order kinetics model. The SFO hypothesizes that the abundance of decaying microorganism in soils is greater than the applied pesticide molecules in soils, hence the rate of degradation at any time is directly proportional to the actual amounts of remaining pesticide in the soils (FOCUS 2006). In other words, for SFO kinetics, the calculated half-life is constant throughout the experimentation, and independent of the initial amount of the pesticide in soils

[b]DFOP (double first-order in parallel) model assumes that two distinct compartments are operating in parallel, each with a specific relative size g or (1−g), and degradation rate constants k_1 and k_2, respectively

during the pot study, i.e. 22 °C, 70% and 600 lx light intensity with a 12/12 h light–dark cycle, respectively, (ii) synergistically accelerated degradation by pepper root exudates, rhizomicroflora and endophytes, and (iii) translocation of fluopyram from root to shoot (Sun et al. 2020).

Rate of Degradation in Soils—Field Studies

According to the reports of JMPR (2010a), DAR (2011d), EFSA (2013) and APVMA (2015), the dissipation of fluopyram was studied in bare soil plots at six European sites (with an application rate of 250 g ai ha^{-1} of fluopyram 250 g L^{-1} SC formulation) and at five USA sites (with an application rate of 500 g ai ha^{-1} of fluopyram 500 g L^{-1} SC). The dissipation results of these studies are summarized in Tables 5 and 6.

The dissipation of fluopyram from 0 to 20 cm soil depth was varied due to different types of European soil (i.e. silt loam, sandy loam and loam) with reported half-lives ranging from 21 to 347–386 days, and estimated DT_{90} values ranging from 487 to > 1000 days. Similar results have been obtained for USA soils, wherein the fluopyram was dissipated with the half-lives ranging from 24 to 537 days, and DT_{75} values ranging from 502 to > 1000 days. Moreover, results of long-term persistence studies with sandy loam (in Germany) and silt loam (in France) soils indicate that even with a single application of fluopyram at 250 g ai ha^{-1}, 29% and 53% residues of initial concentration persisted, respectively, in sandy loam and silt loam soils after a year-long period of

Table 5 The DT_{50} and DT_{90} values for fluopyram 250 SC in European field soils

Country; soil type; soil property	DT_{50} (day)	DT_{90} (day)	Model used
As reported in the DAR (2011a)			
Germany; silt loam; pH 6.9, OC 1.07%	145	> 1000	DFOP
UK; sandy loam; pH .8.1, OC 1.07%	164	> 1000	DFOP
Sweden; loam; pH 8.1, OC 1.38%	386	> 1000	SFO
France; silt loam; pH 7.3, OC 0.79%	318	> 1000	DFOP
Spain; loam; pH 6.7, OC 0.68%	147	487	SFO
Italy; silt loam; pH 8.2, OC 1.21%	21	512	DFOP
As reported in the JMPR (2010a) and EFSA (2013)			
Germany; silt loam; pH 6.9, OC 1.07%	206	684	SFO
	146	> 1000	DFOP
UK; sandy loam; pH .8.1, OC 1.07%	315	> 1000	SFO
	239	> 1000	DFOP
Sweden; loam; pH 8.1, OC 1.38%	312	> 1000	SFO
	179	> 1000	DFOP
France; silt loam; pH 7.3, OC 0.79%	391	> 1000	SFO
	347	> 1000	DFOP
Spain; loam; pH 6.7, OC 0.68%	147	487	SFO
	100	97	DFOP
Italy; silt loam; pH 8.2, OC 1.21%	118	391	SFO
	21.3	512	DFOP

Table 6 Dissipation times for fluopyram 500 SC in USA field soils (JMPR 2010a; DAR 2011a)

Location; soil type; soil property	DT_{50} (day)	DT_{75} (day)	Model used
Washington; Sandy loam; pH 8.1, OM 0.8%	295	603	SFO
	163–166	784–816	DFOP
New York; Loamy sand; pH 6.2, OM 2.2%	677	> 1000	SFO
	537–539	> 1000	DFOP
North Dakota; Loam; pH 7.0, OM 5.8%	382	770	SFO
	83–86.7	> 1000	DFOP
Georgia; Loamy sand; pH 5.9, OM 0.8%	147.5	295	SFO
	24.1	502–521	DFOP
California; Sandy loam; pH 7.8, OM 1.1%	284	578	SFO
	174–175	665–688	DFOP

study (JMPR 2010a; DAR 2011a; APVMA 2015). Whereas with consecutive annual applications, 57% and 59% of initial residues were persisted in the respective soils after two years of studies. These results of bare plot studies conferred the highly persistent nature of fluopyram in soils.

Fluopyram is sprayed on the plant canopy/foliage and is supposed to be deposited primarily on leaf surfaces, with foliar dissipation likely to be the dominant process. However, washing-off due to rain events or any other means and spray drift can result in increased residue loads in the soil. Furthermore, since fluopyram is a systemic ingredient, a portion of the foliar-applied ingredient can translocate to plant roots, allowing residues to enter soils. Besides that, the incorporation of after-harvest crop residues and the falling of plant leaves into the soil can increase soil residue loads. Given that foliar-applied molecules ultimately enter the soil, it is also important to assess the dissipation of soil residues following foliar application. To this, Guan et al. (2012) stated that the fluopyram dissipated rapidly in soils with DT_{50} ranging from 5.8 to 6.2 days after foliar application of fluopyram (500 g L^{-1} SC formulation) at 300 mL ha^{-1} on cucumber plant. Similarly, Dong and Hu (2014) reported a first-order half-life of 15.8 days in sandy loam (pH 6.79, organic matter, OM 1.71%) and 24.8 days in clay loam soil (pH 732, OM 3.89%) after foliar application at the level of 300 g ai ha^{-1} of fluopyram (200 g L^{-1} SC) on watermelon crop. In a separate experiment at two different sites, wherein the watermelon fields were sprayed twice and thrice (at 7-day interval) with fluopyram levels of 200 g and 300 g ai ha^{-1}, fluopyram residues in the soil ranged from 0.0378 to 0.169 mg kg^{-1} at 28 days after the last spray (Dong and Hu 2014). In another study with vegetables grown in greenhouse soil, Wei et al. (2016) reported a constant declined in fluopyram residues with relatively much shorter half-lives of 4.2–5.7 days in sandy loam soil, wherein fluopyram 500 g L^{-1} SC was sprayed at 62.4 g ai ha^{-1} on vegetable crops at the fruit initiation stage. The shorter half-lives were largely due to relatively higher temperature and humidity in the greenhouse ecosystem that could accelerate the dissipation process (Wei et al. 2016). Further, optimum microclimate and hotbed conditions of the greenhouse system are known to proliferate the indigenous rhizomicroflora, epiphytes and endophytes, and which could be synergically degraded the agrochemical rapidly than that of open-field conditions (Wei et al. 2016).

Also, in the dissipation studies carried out in India (Patel et al. 2016; Chawla et al. 2018; Katna et al. 2018; Matadha et al. 2018; Mohapatra et al. 2018; Patil et al. 2018), fluopyram showed lower persistence in soils. After 52 days of foliar spray of fluopyram (i.e. Luna experience 400 g L^{-1} SC; combination pre-mix product of fluopyram 200 g L^{-1} + tebuconazole 200 g L^{-1}) on onion crop, Patel et al. (2016) reported fluopyram residues of 0.07 mg kg^{-1} and 0.12 mg kg^{-1} in sandy loam soils for standard dose (150 g ai ha^{-1}) and double dose (300 g ai ha^{-1}), respectively. In another study on French beans with three foliar applications of fluopyram 200 g L^{-1} SC, at levels of 250 and 500 g ai ha^{-1} (sprayed at the pod development stage and thereafter 10 days of interval), Katna et al. (2018) reported that soil fluopyram residues reached below LOQ of 0.05 mg kg^{-1}at 50 days after last foliar spray. Similar results of soil fluopyram residues below LOQ of 0.01–0.05 mg kg^{-1} and no detection of any of fluopyram metabolites in soil have been reported after four foliar applications of fluopyram 200 g L^{-1} SC at levels of 300 and 600 g ai ha^{-1} on mango tree (Mohapatra et al. 2018), and after three foliar sprays of fluopyram 200 g L^{-1} SC at levels of 75 and 150 g ai ha^{-1} on pomegranate tree (Patil et al. 2018).

However, with an application of fluopyram 400 g L^{-1} SC as a soil drench (at levels of 250 and 500 g ai ha^{-1} doses; two soil drenchings, 1st at the fruit initiation stage and 2nd at 15 days after 1st drenching) to the roots of cucumber plants, Chawla et al. (2018) observed a relatively higher fluopyram residues in soils (0.19 and 0.59 mg kg^{-1}, respectively, for 250 and 500 g ai ha^{-1} doses) at 15 days after 2nd soil drenching. Correspondingly, Matadha et al. (2018) also reported the fluopyram residues of 0.80–1.05 mg kg^{-1} in the soil at 60 days after soil drenching of fluopyram 200 g L^{-1} SC at a level of 0.5 mL L^{-1} of water. By the 60th day of soil drenching, 0.08–0.1 mg kg^{-1} residues of metabolite BZM in soil were detected. Moreover, the dissipation of residues (fluopyram + BZM) in soil followed the first-order kinetics with a half-life of 36 days (Matadha et al. 2018).

According to the above-mentioned in vitro and field studies, fluopyram dissipation varies with soil type, agroclimatic conditions, as well as application method and dose.

Sorption and Leaching of Fluopyram in Soils

Although pesticide applies on the crop or weed foliage for pest/weed/disease control, their residues ultimately ended up in soil through various ways such as it be washed away by rain into soil, translocation into the soil by plant or incorporated into the soil through crop stubble. Once in the soil ecosystem, molecules partition between solid and aqueous phases of soil (so-called sorption/adsorption), and the extent of sorption behaviour affects each aspect of molecules behaviour in soil (Wauchope et al. 2002). The mobility of a pesticide in the soil is determined by the extent and strength of sorption, which is most influenced by various soil physicochemical properties. The existing literature/reports on fluopyram mobility in soil (DAR 2011a; EFSA 2013; VKM 2014; APVMA 2015) indicate that the fluopyram showed moderate soil mobility, while its metabolite 7-hydroxy showed moderate to high soil mobility.

According to the draft assessment report (DAR 2011a), batch equilibrium adsorption/desorption behaviour of fluopyram was studied for five soils (3 European and 2 US soils) over the concentration range of 0.01–1.0 mg ai L^{-1}. Results showed that percentage adsorption of the applied amount was 48–76%, 71–83%, 58–74%, 44–62% and 55–74% in sandy loam, silt loam, loam, loamy sand and clay loam soils, respectively; and the adsorption coefficient $K_{d(ads)}$ ranged from 3.16 to 8.37 mL g^{-1} and the apparent adsorption constant $K_{OC(ads)}$ ranged from 266 to 460 mL g^{-1}. There are two well-accepted classifications for chemical mobility in soil (McCall et al. 1981; FAO 2000b) based on adsorption coefficient, K_{OC}. According to McCall's classification, a chemical can be categorized as very high, high, medium, low, slightly and immobile, respectively, for $K_{OC(ads)}$ values 0–50, 50–150, 150–500, 500–2000, 2000–5000 and > 5000. Similarly, as per FAO classification, it is classified as highly mobile, mobile, moderately mobile, slightly mobile, hardly mobile and immobile, respectively, for the $K_{OC(ads)}$ values < 10, 10–100, 100–1000, 1000–10,000, 10,000–100,000 and > 10,000. Based on the reported $K_{OC(ads)}$ values, the fluopyram can be classified as having medium or moderate mobility in all tested soils. Moreover, higher fluopyram desorption coefficients were observed with $K_{d(des)}$ ranging from 6.32 to 13.15 mL g^{-1} and $K_{OC(des)}$ ranging from 444 to 834 mL g^{-1}, indicating a strong binding of fluopyram once adsorbed to the soil (DAR 2011a; APVMA 2015).

According to the other reports (EFSA 2013; VKM 2014), adsorption of fluopyram can also be classified as medium or moderate mobile, with Freundlich adsorption coefficients $K_{F(ads)}$ values ranged from 2.94 to 6.82 mL g^{-1} and $K_{OC(ads)}$ ranged from 233 to 400 mL g^{-1}. Whereas, the adsorption of metabolite fluopyram-7-hydroxy can be classified as high or mobile to moderately mobile, with the $K_{F(ads)}$ 0.99–2.39 mL g^{-1} and $K_{oc(ads)}$ 85–149 mL g^{-1}. The desorption coefficients, $K_{d(des)}$ and $K_{OC(des)}$, of fluopyram-7-hydroxy were 3.38–5.97 mL g^{-1} and 237–373 mL g^{-1}, respectively (VKM 2014). In another report (NYSDEC 2017), the adsorption coefficient (K_{OC}) of fluopyram ranged from 316 to 591 mL g^{-1}, indicating moderate mobility of fluopyram in certain soils.

Overall, fluopyram is moderately mobile within the soil and can consequently be expected to occur in surface water runoff and/or in groundwater (EPA 2012). The majority of the studies found quantifiable residues of fluopyram in the top 20–30 cm soil layers with occasional detections at soil depths greater than 30 cm (JMPR 2010a; DAR 2011a; APVMA 2015). Furthermore, no transformation product was detected beyond 30 cm soil depth, indicating that the metabolites have a low potential to leach and contaminate the groundwater (DAR 2011a).

Also with relatively low water solubility, 10% and 2% of initial fluopyram concentrations (fluopyram 500 g L^{-1} SC applied at the level of 500 g ai ha^{-1} to bare sandy loam soils; at California and Washington sites) were detected, respectively, in the 30–60 and 60–90 cm soil depths after 2 years of studies, which may be attributed to the excessive irrigation practices (JMPR 2010a).

Another document (NYSDEC 2017) also reported fluopyram residues at 75–90 cm soil depth at a concentration of 3.04 µg kg^{-1} after 665 days, following a single application of fluopyram at a level of 500 g ai ha^{-1}. According to the LEACHP (Leaching Estimation And Chemistry Pesticide) modelling, fluopyram could leach from the Riverhead soil profile (i.e. characterized by deep, well-drained, moderately coarse-textured soils) at the maximum concentrations of 3.56–18.20 µg kg^{-1} depending on half-lives and application rates (NYSDEC 2017).

Dissipation of Fluopyram in Water

Abiotic Transformation: Hydrolysis and Photolysis

Fluopyram can reach aquatic systems through spray drift, overland runoff or through the movement of soil particles containing bound residues. As per the pesticide residues and assessment reports (JMPR 2010a; DAR 2011a; EFSA 2013; VKM 2014; APVMA 2015), fluopyram is hydrolytically stable at all environmentally relevant pH conditions and no major metabolites have been detected in aqueous and water–sediment systems.

The hydrolysis of phenyl-UL-^{14}C-fluopyram was studied at 1.0 mg L^{-1} in the dark at 50 °C in sterile buffer solutions at pH 4, pH 7 and pH 9 for 5 days (JMPR 2010a; DAR 2011a; ECHA 2013), and neither hydrolysis was observed under acidic, neutral and alkaline conditions, nor were major degradation products detected.

Since fluopyram does not absorb light at wavelengths greater than 292 nm, direct photolysis in an aqueous solution has little effect on the total transformation of fluopyram in the environment (DAR 2011a; APVMA 2015). Similarly, when fluopyram (1.0 mg L^{-1}) was exposed to continuous artificial sunlight of > 290 nm for 13 days, it underwent the least transition to fluopyram-lactam by indirect photolysis in sterile aqueous buffer solution (pH 7.0). Fluopyram residues were reduced from 99.5 to 64% of phenyl label, and from 100 to 72% of the applied pyridyl label after 13 days of continuous artificial sunlight in sterile aqueous buffer solutions (pH 7) at 25 °C. The estimated DT_{50} and DT_{90} values were 21–25 (mean 23 days) and 70–83 days, respectively, based on simple first-order kinetics (JMPR 2010a; DAR 2011a; ECHA 2013). Based on the estimated mean DT_{50} of 23 days in aqueous buffer solutions, the predicted environmental DT_{50} was 57 summer days in Phoenix, US and 89 summer days in Athens, Greece (JMPR 2010a; DAR 2011a; ECHA 2013).

Fluopyram in natural water under non-sterile conditions was photodegraded with a DT_{50} of 21 days while exposed to continuous irradiation (JMPR 2010a; DAR 2011a; ECHA 2013). This estimated DT_{50} was equivalent to 87 summer days in Phoenix, US and 135 summer days in Athens, Greece. Unlike sterile buffer solutions, no transformation products had been observed in the natural water. These results imply that photolysis would not be the most important pathway of degradation in a natural aquatic environment. However, photodegradation of fluopyram under simulated UV ($\lambda \geq 200$ mm) and sunlight ($\lambda \geq 290$ mm) irradiations was found to be faster in a neutral solution compared to acidic and alkaline solutions (Dong and Hu 2016). The author attributed the decreased rate of photodegradation in acidic media to the presence of chloride ions and absorbance of UV irradiation at 254 nm by acetic acid in acidic media; while in alkaline media to the deactivation of hydroxyl radicals and formation of O^- ions. Further, the presence of fulvic acid, nitrate, Fe^{+3} and titanium dioxides (TiO_2) slightly enhanced or decreased the photodegradation of fluopyram under UV irradiation, wherein the presence of 5 mg Fe^{+3} L^{-1} and 500 mg TiO_2 L^{-1}, the photodegradation rates under simulated sunlight irradiation were increased, respectively, by sevenfold and threefold faster (Dong and Hu 2016). Titanium dioxide as a photocatalyst produces strongly oxidizing species (OH^-, H^+, O_2^-, HOO radicals) which degrades organic compounds, whereas the organic ligands in fluopyram molecule (such as carbonyl, pyridine and N–H bonds) might form Fe^{+3}-fluopyram complex, which would accelerate the fluopyram degradation (Dong and Hu 2016). In addition, the author also proposed a degradation pathway of fluopyram in water, identifying three transformation products: (i) fluopyram-lactum, formed by intramolecular eliminate of HCl; (ii) *N*-(2-[3-hydroxy-5-(trifluoromethyl)-2-pyridinyl]ethyl)-2-(trifluoromethyl)benzamide, formed by hydroxyl substitution and (iii) *N*-(2-[5-trifluoromethyl)-2-pyridinyl]ethyl)-2-(trifluoromethyl)benzamide, formed by hydrogen extraction. Later in another study (Hu et al. 2019), the author reported six photolytic transformation products of fluopyram in aqueous TiO_2 suspension, which were triggered by the hydroxyl substitution of Cl atom, intramolecular cyclization, cleavage of amido bond, loss of trifluorotoluene and attack of the hydroxyl group on the benzene ring.

Fluopyram molecule comprises six fluorine atoms, which can decompose into toxic fluorine-containing compounds, and thus likely to contaminate the environment. To this, detoxification/degradation of fluopyram in water was studied by Li et al. (2020a) by employing ozone, microbubble and ozone-microbubble (OMB) treatments, and results revealed that the OMB treatment had efficiently degraded fluopyram with a half-life of 0.28 h, and 60–80% of fluopyram concentration (2.5, 5, and 10 mg L^{-1}) was decomposed due to OMB treatment for 60 min. The author proposed two degradation mechanisms in water under OMB treatment; the first was the direct dichlorination and hydroxyl substitution of the C–Cl bond and the second was cleavage and oxidation of amide bond.

Biotransformation: Degradation Pathways in Water–Sediment

Fluopyram considerably partitioned from the water to the sediments in aerobic water–sediment system. According to the aerobic degradation study of fluopyram in two pond water–sediment systems, the dissipation of fluopyram in the water phase was found to be medium (with DT_{50} of 14–26 days), while the dissipation in the total water–sediment system was found to be low (with $DT_{50} > 648$ days). These values showed the fluopyram is more persistent in the water–sediment environment (DAR 2011a; ECHA 2013; APVMA 2015). Anaerobic biotransformation study of phenyl- and pyridyl-labelled fluopyram in a pond water–sediment system have also been discussed in these reports, with no transformation products detected and limited mineralization of fluopyram observed by the end of the study. The DT_{50} and DT_{90} values for the entire water–sediment system were observed over 1000 days, demonstrating a strong persistent nature of fluopyram in the aquatic system under anaerobic conditions. Thus, neither aerobic nor anaerobic biodegradation would be a typical degradation route for fluopyram in the water–sediment ecosystem.

Dissipation of Fluopyram in Plants

Various publications (JMPR 2010a; DAR 2011d; EFSA 2013; APVMA 2015) have addressed studies on metabolism, distribution and fluopyram residues in different plant produce (in grapes, potatoes and beans after foliar sprays of radio-labelled fluopyram at recommended rates; in red bell pepper after application of radio-labelled fluopyram through drip irrigation under greenhouse condition). Results show that after 4–51 days of last foliar spray, fluopyram constitutes the major component of radioactive residues, accounting for > 85% total radioactive residues (TRR) in grapes, potato leaves and bean leaves. However, the fluopyram was observed in lower proportions in potato tubers, drip-irrigated peppers, dry and succulent beans (where the commodities were not directly exposed to fluopyram sprays), representing 5–21% TRR. Residues in these plant matrices were largely composed of the metabolites resulting from cleavage of the parent molecule, such as BZM, PAA and PCA. Moreover, a similar metabolic pathway was found in drip-irrigated pepper, with fluopyram, PCA and PAA-glycosides accounting for 16–44% TRR in fruits. The residues of fluopyram in rotational crops (wheat, Swiss chard and turnips) were also investigated (JMPR 2010a; DAR 2011d; EFSA 2013;

APVMA 2015). The metabolic pathway was found to be similar, with parent fluopyram being identified as a major component of the residues (20–94% TRR in all plant parts analysed), and in addition, metabolites viz., BZM, fluopyram-benzoic acid, PCA and fluopyram-methyl sulfoxide being detected at significant levels in wheat grains. The authors have discovered that hydroxylated metabolites and their conjugates were present in much higher amounts in rotational crops than in primary crops.

Overall, as shown in Fig. 3, the metabolic pathway in plants (grape, potato, bean, red bell pepper, apple) consists of (i) hydroxylation of fluopyram leading to metabolites, 7-hydroxy and 8-hydroxy; (ii) conjugation of hydroxylated metabolites with glucose, malonic acid, glucoside and glucuronic acid; (iii) cleavage of the hydroxylated fluopyram and subsequent oxidation leading to metabolite, BZM and fluopyram-benzoic acid from the phenyl moiety, while metabolites PAA, fluopyram-methyl sulfoxide and PCA from the pyridyl moiety of the active substance.

Residue trials conducted throughout the United States and Canada using end-use products containing fluopyram in/on various fruits and vegetable crops were enough to support the proposed Canadian and USA maximum residue limits (Health Canada PMRA 2014). The Dissipation and persistence of fluopyram for different vegetable crops have also been studied in India and China (Table 7). It is evident from the data presented in Table 7 that fluopyram residues were reached below quantification or maximum residue limits, and the pre-harvest interval (PHI) for different plants ranged between 1 and 38 days. The persistence of fluopyram in different vegetable and fruit crops have been studied at Anand centre of Anand Agricultural University as a part of the All India Network Project on pesticide residues (AINP 2016a,b,c,d), and the findings are summarized in Table 7.

Conclusion

This paper aims to put together all existing published scientific literature on new SDHIs fungicide, fluopyram, to provide a broader viewpoint on its environmental fate. Based on the presented literature review of recent scientific studies, it can be concluded that (i) The fluopyram is degraded slowly in most studied arable soils and its dissipation is conformed to either simple first-order or double first-order in parallel kinetics; (ii) The fluopyram is moderate to highly persistent in soils with half-lives ranging from 162 to 746 days in laboratory aerobic studies and 21–539 days in bare soil dissipation under real-field conditions; (iii) According to the reported adsorption coefficients, fluopyram can be categorized as 'medium or moderately mobile' in the soil, and fluopyram residues have been found in soil depths > 30 cm in some of the field studies, so fluopyram can leach and contaminate groundwater depending on the types of soil and location; (iv) The fluopyram residues in plant parts or finished products are typically found to be below either quantification or maximum residue limit in most studies, with pre-harvest intervals varying from 1 to 38 days.

Since fluopyram is a persistent compound that can build up in the soil after repetitive treatments, and because of the possible uptake of such aged soil residues in rotational crops, further studies are needed to understand the environmental fate of fluopyram and their metabolites in the soil–plant system. Only a few studies have looked at the effects of fluopyram on soil microbes, but the findings are quite inconsistent. Thus, further rigorous scientific studies on the impact of fluopyram on soil biological properties are needed to ascertain its ecotoxicological effects and consequences for soil health. As of now, only two studies in the literature (Sjerps et al. 2019; Pinasseau et al. 2020) reported the occurrence of fluopyram residues in groundwater or drinking water source. In future research, therefore it will be important to quantify the fate processes such as sorption/desorption, mobility and leaching of fluopyram that occur in the soil profile and unsaturated zone. The findings of such studies would aid in improving the accuracy of various models used to estimate the concentrations of fluopyram in the subsurface and groundwater. According to toxicity predictions made by Hu et al. (2019), the photolytic degradation products of fluopyram are potentially more toxic than fluopyram, and therefore a detailed ecotoxicological study of such degraded products is needed to get a true risk assessment.

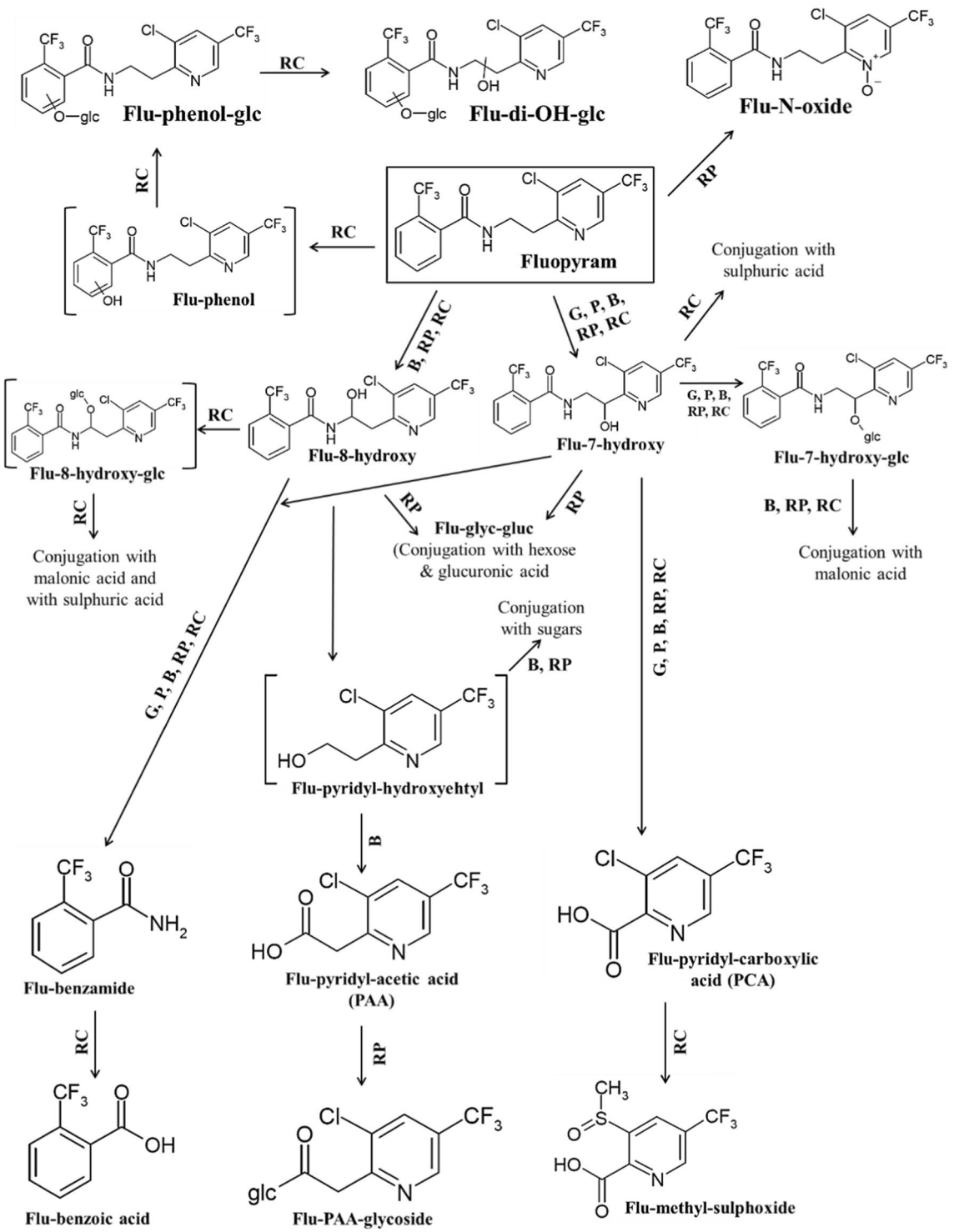


Fig. 3 Metabolic pathways of fluopyram in plants (JMPR 2010a; DAR 2011d). Metabolites in square brackets [] are postulated intermediates; *Flu* Fluopyram, *glc* conjugate with glucose, *glyc* conjugate with glycose, *G* grapes, *P* potatoes, *B* beans, *RP* red pepper, *RC* rotational crops

Table 7 Dissipation and persistence of fluopyram residues in plants

Test crop(s)	Formulation; Application rate	Findings	References
Foliar spray of fluopyram			
Grapes, tomato strawberry	500 g L^{-1} SC; 250 g ai ha^{-1}	PHI: 1–21 days	DAR (2011e)
Cucumber	500 g L^{-1} SC; 300 mL ha^{-1}	DT_{50}: 3.7–4.3 days	Guan et al. (2012)
Watermelon	200 g L^{-1} SC; 200 and 300 g ai ha^{-1}	DT_{50}: 6.4–6.6 days PHI: 7 days	Dong and Hu (2014)
Onion & spring onion	200 g L^{-1} SC; 75 and 150 g ai ha^{-1}	DT_{50}: 8.8–9.1 days PHI: 7 days	Patel et al. (2016)
Chilli	200 g L^{-1} SC; 250 and 500 g ai ha^{-1}	DT_{50}: 1.16–1.21 days PHI: 1 day	Saha et al. (2016)
Tomato, pepper, cucumber	500 g L^{-1} SC; 62.4 g ai L ha^{-1}	DT_{50}: 5.7 days PHI: 7–21 days	Wei et al. (2016)
Tomato	200 g L^{-1} SC; 375 g ai ha^{-1}	DT_{50}: 2.8–12.0 days PHI: 3 days	Hussan et al. (2017)
Apple	200 g L^{-1} SC; 750 g ai ha^{-1}	DT_{50}: 7.7–10.5 days PHI: 3 days	Podbielska et al. (2017)
French bean	200 g L^{-1} SC; 150 and 300 g ai ha^{-1}	DT_{50}: 3.8–3.9 days PHI: 10 days	Katna et al. (2018)
Mango	200 g L^{-1} SC; 125 and 250 g ai ha^{-1}	DT_{50}: 4.3–5.7 days PHI: 28–38 days	Mohapatra et al. (2018)
Pomegranate	200 g L^{-1} SC; 75 and 150 g ai ha^{-1}	DT_{50}: 4.0–5.4 days PHI: 7–14 days	Patil et al. (2018)
Melon	250 g L^{-1} SC; 112.5 g ai ha^{-1}	DT_{50}: 4.5–6.2 days PHI: 3–10 days	Yizhi et al. (2020)
Chilli	200 g L^{-1} SC; 100 and 200 g ai ha^{-1} (3 sprays)	Residues detected below LOQ of 0.05 mg kg^{-1}. DT_{50}: 5.6–5.7 days	ANIP (2016a)
Mango	0.075% and 0.15% ai per tree (4 sprays)	Residues detected below LOQ of 0.05 mg kg^{-1}. DT_{50}: 1.9–10 days	ANIP (2016d)
Soil drenching of fluopyram and dripping in irrigation water			
Cucumber	200 g ai ha^{-1} SC; 250 and 500 g ai ha^{-1}	Residues detected below LOQ of 0.05 mg kg^{-1}. PHI: 15 days	Chawla et al. (2018)
Tomato, bell pepper	17.7% SC; 0.5 mL L^{-1}	Residues detected < MRLs	Matadha et al. (2018)
Cherry tomato, Cucumber	500 g L^{-1} SC; 625 and 1250 g ai ha^{-1}; through irrigation water	The behaviour of fluopyram does not fit any type of kinetic classical model of degradation	Vargas-Pérez et al. (2020)
Banana	400 g L^{-1} SC;625 and 1250 g ai ha^{-1} (single application); 250 and 500 g ai ha^{-1} (twice applications)	Residues detected below LOQ of 0.05 mg kg^{-1}. DT_{50}: 5.74–34.6 days (in banana leaves)	ANIP (2016b)
Tomato	400 g L^{-1} SC; 250 and 500 g ai ha^{-1} (twice applications)	Residue detected below LOQ of 0.05 mg kg^{-1}	ANIP (2016c)

Fluopyram 500, 400, 250, 200 g L^{-1} SC are equivalent to the 41.7, 34.4, 21.4 and 17.5% concentration of fluopyram, respectively

DT_{50} half-live, *PHI* pre-harvest interval

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Data availability Data transparency.

Code availability Software application or custom code.

Declarations

Conflict of interest Authors have no conflict of interest.

Humans and animals rights Additional declarations for articles in life science journals that report the results of studies involving humans and/or animals.

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