Local-scale dynamics of plant-pesticide interactions in a northern Brittany agricultural landscape

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Authors :

Anne-Antonella Serra[a], Anne-Kristel Bittebière[b], Cendrine Mony[a], Kahina Slimani[a], Frédérique Pallois[a], David Renault[a], Ivan Couée[a], Gwenola Gouesbet[a], Cécile Sulmon[a]

Affiliation :

[a] Univ Rennes, Université de Rennes 1, CNRS

ECOBIO [(Ecosystems-Biodiversity-Evolution)] - UMR 6553

Campus de Beaulieu

263 avenue du Général Leclerc

F-35042 Rennes Cedex, France

[b] Université de Lyon 1, CNRS

UMR 5023 LEHNA

43 Boulevard du 11 novembre 1918,

F-69622 Villeurbanne Cedex, France

Corresponding author :

Ivan Couée

Univ Rennes, Université de Rennes 1, CNRS

ECOBIO [(Ecosystems-Biodiversity-Evolution)] - UMR 6553

Campus de Beaulieu

263 avenue du Général Leclerc

F-35042 Rennes Cedex, France

Phone : 33-223235123, Fax : 33-223235026

E-mail: Ivan.Couee@univ-rennes1.fr

Abstract

Soil pollution by anthropogenic chemicals is a major concern for sustainability of crop production and of ecosystem functions mediated by natural plant biodiversity. Understanding the complex effects of soil pollution requires multi-level and multi-scale approaches. Non-target and agri-environmental plant communities of field margins and vegetative filter strips are confronted with agricultural xenobiotics through soil contamination, drift, run-off and leaching events that result from chemical applications. Plant-pesticide dynamics in vegetative filter strips was studied at field scale in the agricultural landscape of a long-term ecological research network in northern Brittany (France). Vegetative filter strips effected significant pesticide abatement between the field and riparian compartments. However, comparison of pesticide usage modalities and soil chemical analysis revealed the extent and complexity of pesticide persistence in fields and vegetative filter strips, and suggested the contribution of multiple sources (yearly carry-over, interannual persistence, landscape-scale contamination). In order to determine the impact of such persistence, plant dynamics was followed in experimentally-designed vegetative filter strips of identical initial composition (*Agrostis stolonifera*, *Anthemis tinctoria* / *Cota tinctoria, Centaurea cyanus*, *Fagopyrum esculentum*, *Festuca rubra*, *Lolium perenne*, *Lotus corniculatus*, *Phleum pratense*, *Trifolium pratense*). After homogeneous vegetation establishment, experimental vegetative filter strips underwent rapid changes within the following two years, with *Agrostis stolonifera*, *Festuca rubra*, *Lolium perenne* and *Phleum pratense* becoming dominant and with the establishment of spontaneous vegetation. Co-inertia analysis showed that plant dynamics and soil residual pesticides could be significantly correlated, with the triazole fungicide epoxiconazole, the imidazole fungicide prochloraz and the neonicotinoid insecticide thiamethoxam as strong drivers of the correlation. However, the correlation was vegetative-filter-strip-specific, thus showing that correlation between plant dynamics and soil pesticides likely involved additional

factors, such as threshold levels of residual pesticides. This situation of complex interactions between plants and soil contamination is further discussed in terms of agronomical, environmental and health issues.

Keywords :

Ecotoxicity, Persistent Organic Pollutants, Pesticides, Plant community dynamics, Soil pollution, Vegetative filter strips

Highlights :

- Characterization of soil pesticide contamination requires multi-level studies.
- Plant-pesticide dynamics was studied at field scale in vegetative filter strips.
- Pesticide persistence in soils was interannual, multi-source and multi-compound.
- Pesticide persistence and plant dynamics showed case-by-case relationships.
- Epoxiconazole was a strong driver of contamination and vegetation dynamics.

1. Introduction

Soil pollution by anthropogenic chemicals is a major concern for the sustainability of crop production and of ecosystem functions mediated by natural plant biodiversity (Arias-Estévez et al., 2008; MacLeod et al., 2014; Persson et al., 2013; Rodríguez-Eugenio et al., 2018; Virto et al., 2015). Understanding the range of soil pollution effects on plants (Rodríguez-Eugenio et al., 2018) requires multi-level and multi-scale approaches from plant cells (Alberto et al., 2017; Arias-Estévez et al., 2016; Vivancos et al., 2011) to landscapes, countries and continents (Billeter et al., 2008; Fried et al., 2018; Geiger et al., 2010; Liira et al., 2008).

In this context of environmental contamination, vegetative filter strips (VFS), defined as areas planted with grasses or non-crop vegetation, are a best management practice intended to reduce agricultural runoffs into riparian zones and surface water (Carluer et al., 2017; Dillaha et al., 1989; Gene et al., 2019; Krutz et al., 2005; Magette et al., 1989; Serra et al., 2016). Agri-environmental plant communities in VFS are necessarily confronted with agricultural pollutants through residual soil contamination, drift, run-off and leaching events that result from chemical applications (Gove et al., 2007; Helander et al., 2012; Rodríguez-Eugenio et al., 2018). Confrontation of these plant communities with pesticides and other agricultural contaminants is designed to mitigate the dispersion and the impact of pollutants in terrestrial and aquatic environments (Collins et al., 2014; Krutz et al., 2005; Serra et al., 2016; Stehle et al., 2011). VFS plant communities, like field margin plant communities (Fried et al., 2018), therefore constitute a useful ecological system for analysing plant-pesticide interactions.

VFS plants are confronted with contrasted environments, with dense crops and in-field treatments (pesticides, fertilizers, watering) on one side and with wild plant communities, heterogeneity and various degrees of canopy openness on the other side. Thus, in addition to the physico-chemical properties of the environment (soil, climate, hydrology), all of these contrasted factors can interfere with natural plant community evolution and have an impact on plant dynamics in VFSs or field margins (Boutin et al., 2019; De Snoo and Van Der Poll, 1999; Kleijn and Snoeijing, 1997; Kleijn and Verbeek, 2000; Pellissier et al., 2014). It is however difficult to discriminate between the effects of these different factors and to demonstrate correlations with a particular factor. For instance, in their analysis of the correlations between a very large panel of field margin vegetation traits and farming practice parameters, Fried et al. (2018) state that "contrary to our expectations, we did not find any particular trait associated with more intensive use of herbicides".

Moreover, plant-pesticide interactions and the impact of pesticide pollution on plant dynamics have been mainly characterised at the widely-diverging scales of experimental exposure of specific plant species (Brown et al., 2009; Peterson et al., 1994; Serra et al., 2019) and of landscape-level correlations between plant communities and pesticide usage (Billeter et al., 2008; Fried et al., 2018; Geiger et al. 2010). At the same time, the emphasis of numerous studies on the impacts of glyphosate and AMPA (Hénault-Ethier et al., 2017; Primost et al., 2017; Silva et al., 2018; Van Bruggen et al., 2018) does not reflect the true variety of pesticide applications in European landscapes, and may overlook other chemical threats (Satapute et al., 2019; Serra et al., 2019; Zubrod et al., 2019). Global parameters of pesticide applications, such as treatment frequency index (Fried et al., 2018), may not be informative enough, and may have to be complemented with actual measurements of pesticide environmental levels. Studies at field level are therefore necessary to assess how the diversity and complexity of pesticide use and the variety of agricultural systems, landscapes and climates (Helander et al., 2012; Krutz et al., 2005; Serra et al., 2016) influence plant-pesticide interactions, and must be integrated in risk assessments for agronomy, environment and health on a regional scale.

In the present study, in order to complement global country- or continent-scale approaches (Billeter et al., 2008; Fried et al., 2018; Geiger et al., 2010; Larroudé et al., 2013; Liira et al., 2008), field-scale analysis of VFSs was carried out in a specific bocage agricultural landscape within a geographical region (Brittany, northwestern France) showing agricultural and ecological contrasts. Brittany (Fig. 1), with a smooth relief and a long coastline along the Atlantic Ocean, the Celtic Sea and the English Channel, is a remarkable peninsula of Central Western Europe (Virto et al., 2015), as well as a major agricultural region of France, especially in terms of animal farming for milk and meat, maize cultivation, and vegetable crops (Piel et al., 2012). The regional landscape is thus dominated by intensive agriculture and livestock activities, which have led to nutrient and xenobiotic contaminations in the atmosphere (Bedos et al., 2002), field margin soils (Serra et al., 2013), surface waters (Piel et al., 2012), and estuaries (Monbet, 2004). At the same time, the bocage structure provides a high level of heterogeneity, with positive effects on biodiversity and ecosystem services (Alignier and Aviron, 2017; Gil-Tena et al., 2015). In this contrasted agricultural context, farm- and field-scale analysis of pesticide usage and environmental dissemination was carried out in order to determine to what extent pesticide-related factors could be primary or secondary drivers of the dynamics of non-target plants in VFSs.

2. Materials and methods

2.1. Study area and set-up of experimental VFSs

The sites of study consisted in a set of five distinct fields and associated VFSs located in the bocage area (48° 36' N, 1° 32' W, Pleine-Fougères, Ille-et-Vilaine, Brittany, France) of the Zone Atelier Armorique Long-Term Ecological Research (LTER) network (Fig. 1) (Alignier, 2018; Alignier and Aviron, 2017; Gil-Tena et al., 2015). The intensive agriculture of the area involves animal farming and cultivation of maize, wheat, barley, canola and sunflower. The LTER structure, which implies regular and active collaboration with local farmers and local authorities, can give rise to experimental approaches embedded within real-life agriculture, thus integrating interannual observations and local-scale data on agricultural practices. In collaboration with local farmers, the VFS were experimentally designed and initiated in 2010 by sowing a set of nine plant species that are commonly found in field margins, VFS or rotational fallows in European agricultural landscapes (Billeter et al., 2008; Kuussaari et al., 2011; Liira et al., 2008; Ma and Herzon, 2014; Serra et al., 2016; Stehle et al., 2011; Toivonen et al., 2013): bird's-foot trefoil (*Lotus corniculatus*, Fabaceae, dicotyledon), common buckwheat (*Fagopyrum esculentum*, Polygonaceae, dicotyledon), cornflower (*Centaurea cyanus*, Asteraceae, dicotyledon), creeping bentgrass (*Agrostis stolonifera*, Poaceae, monocotyledon), English ryegrass (*Lolium perenne*, Poaceae, monocotyledon), red fescue (*Festuca rubra*, Poaceae, monocotyledon), timothy grass (*Phleum pratense*, Poaceae, monocotyledon), white Dutch clover (*Trifolium pratense*, Fabaceae, dicotyledon), yellow chamomile (*Anthemis tinctoria* / *Cota tinctoria*, Asteraceae, dicotyledon). Dicots and Poaceae were respectively sown at densities of 4 $g.m^{-2}$ and 2.7 $g.m^{-2}$, with equal proportions of plant species in the dicot mixture, and with respective weight/weight proportions of 2%, 15%, 37% and 46% of *Agrostis stolonifera*, *Phleum pratense, Festuca rubra*, and *Lolium perenne*, in the Poaceae mixture. Seeds [*Agrostis stolonifera* (Penncross variety), *Anthemis tinctoria* / *Cota tinctoria* (bulk seeds), *Centaurea cyanus* (bulk seeds), *Fagopyrum esculentum* (bulk seeds), *Festuca rubra* (Herald variety), *Lolium perenne* (Brio variety), *Lotus corniculatus* (Leo variety), *Phleum pratense* (Kaba variety), *Trifolium pratense* (Violetta variety)] were obtained from the Phytosem (Gap, Hautes-Alpes, France) seed company. The five experimental VFSs (VFS-91, VFS-109, VFS-112, VFS-113, VFS-114) were each located between their respective adjacent field and their respective riparian zone of neighbouring streams (Fig. 1), with widths of 4.7 m (VFS-109), 6.1 m (VFS-91, VFS-113, VFS-114) and 9.6 m (VFS-112). Adjacent fields of the VFSs covered areas of 1 ha (VFS-91), 1.5 ha (VFS-109), 2.5 ha (VFS-112), 8 ha (VFS-113) and 10 ha (VFS-114), with field-VFS slopes of 4% (VFS-91), 1%

(VFS-109), 3% (VFS-112), 7% (VFS-113) and 7% (VFS-114). Proximal, central and distal sub-strips relatively to the cultivated field were respectively identified as A, B and C (Fig. 1).

2.2. Agricultural and pesticide usage data

Fields and VFSs of the study area were under the responsibility of local farmers. The five fields were cultivated with maize and wheat. Pesticide treatments (fungicides, herbicides, insecticides) were carried out between the beginning of February and the end of May/beginning of June. VFS vegetation was mowed down at the end of the Summer. Data on pesticide usage (nature of commercial products, methods of application, dates of application, applied dosage) were obtained from the farmers in charge of the different fields. Corresponding levels of active ingredient applications were derived from commercially-available characteristics of the products (Table 1).

2.3. Soil sampling

Soil samples were collected in 2011 and 2012 in sub-strip A and sub-strip C (Fig. 1) of each VFS and in its adjacent field (at a distance of 10 m from the VFS) along five transects that were positioned transversally to the VFS, parallel to each other and at 20 m from each other (Fig. 1). Surface cores (0–30 cm) of approximately 200 g were collected with a manual auger. Soil samples were kept on ice for transport from the study area to the laboratory. After drying at 40°C during 5 days, samples were sieved with a 2-mm sieve and kept frozen at −20°C until chemical determinations.

2.4. Analysis of pesticides

Pesticides were analysed and quantified by different methods that were adjusted to their physico-chemical properties. Extraction and purification of pesticides from soil samples were carried out by water-acetonitrile, essentially as described by Vryzas and Papadopoulou-Mourdikou (2002), and by solid phase extraction on Oasis® HLB cartridges, essentially as described by Dias and Poole (2002). Purified extracts were dissolved in ethyl acetate, evaporated to dryness, and stored at -20°C until analysis. A set of 22 pesticides (Table 2) was analysed by tandem liquid chromatography and mass spectrometry (LC-MS/MS) at the Rovaltain Research Company (Valence, France). A set of 12 pesticides (diflufenican, dimethenamid-P, S-metolachlor, acetochlor, pendimethalin, prochloraz, thiamethoxam, flufenacet, cypermethrin, epoxiconazole, metconazole, prothioconazole) was analysed by tandem gas chromatography and mass spectrometry (GC-MS). The GC/MS system consisted of a Trace GC Ultra chromatograph and a Trace DSQII quadrupole mass spectrometer (Thermo Fisher Scientific Inc., Waltham, MA, USA). A 30 m fused silica column (95% dimethyl siloxane, 5% phenyl polysilphenylene-siloxane, v/v) was used with helium as the carrier gas at a rate of 1 $ml.min^{-1}$. A 1 µl aliquot of each sample was injected using the split mode (25:1). The injector temperature was held at 250 °C, and temperature gradients were as follows: (i) 10 min at 50°C, (ii) from 50°C to 200°C at $15°C.min^{-1}$, (iii) 1 min at 200°C and then from 200°C to 280°C at $8°C.min^{-1}$, (iv) 10 min at 280°C. The MS transfer line was set at 250 °C. Detection was achieved using electron impact ionization. Peaks were accurately annotated using both mass spectra (two specific ions) and retention times. Calibration curves were established by using samples made up of pure reference compounds. Quantification was carried out with the Xcalibur 2.0.7 software (Thermo Fisher Scientific Inc., Waltham, MA, USA).

2.5. Survey of vegetation changes in the VFSs

VFS were botanically surveyed for the observation of the nine initial plant species and of additional spontaneous vegetation over the course of three years (2010, 2011, 2012). Vegetation cover composition was assessed in the 3 different sub-strips of the VFSs (Fig. 1) within 0.25 m² quadrates disposed along five transects (Fig. 1), which were positioned transversally to the VFS, parallel to each other, and at 20 m from each other. The occurrence and the surface cover of each of the nine initial plant species, of additional spontaneous vegetation, and of bare ground were estimated and recorded within all quadrates to assess plant dynamics in the sub-strips of the VFSs. Additional spontaneous vegetation was considered as a whole, and not species by species. Botanical surveys and cover assessment were carried out in a non-destructive manner at the same positions of the quadrates along the 5 transects (Fig. 1) from year to year.*2.6. Calculations and statistical analyses*

Quantitative analysis of soil pesticide levels (Fig. 2-5) was carried out in 5 independent soil samples corresponding to the 5 transects of sampling (Fig. 1). Results were given as the mean (± SEM) of these determinations. Whenever necessary for the calculation of indicative estimations, conversions of pesticide levels relatively to dry weight, fresh weight or volume were carried out with average soil characteristics [bulk density: 1.4 $kg.L^{-1}$; soil moisture: 20% (w/w)] that are found in northern Brittany agrosystems (Binet et al., 2006; Sulmon et al., 2007). Quantitative analysis of plant surface covers (Fig. 3-5) was carried out in 5 quadrates corresponding to the 5 transects of observation (Fig. 1). The means (± SEM) of these determinations were used for further analysis. Pairwise comparisons of means was carried out using the non-parametric Mann–Whitney–Wilcoxon test with the R version 3.1.3 software. In order to test relationships between plant parameters, VFS parameters and pesticide parameters, principal component analysis (PCA) and factorial correspondence analysis (FCA) based on the correlation matrix were carried out using the FactoMineR package of R. FCA breaks down the relationships between sets of variables in a multidimensional way, where each dimension attracts a given amount of information called the inertia. Co-inertia analysis and calculation of RV coefficients (Robert and Escoufier, 1976) were carried out to analyse the relationships between soil residual pesticide and plant surface cover datasets. Co-inertia analysis is used to identify common trends or relationships in the coupling of dataset tables.

3. Results

3.1 Agricultural pesticide environment of the experimental VFSs

The adjacent fields of the 5 VFSs under study (Fig. 1) were cultivated with maize or wheat over the 2010-2012 period. The chemical treatments associated with these crops (Table 1) involved 28 different pesticides, including: (i) herbicides, such as the chloroacetanilides S-metolachlor and acetochlor, (ii) fungicides, such as the imidazole prochloraz, and the triazoles epoxiconazole, metconazole and prothioconazole, (iii) insecticides, such as the pyrethroid cypermethrin and the neonicotinoid thiamethoxam. It must be noted that, besides differences of regulations throughout the world, the regulatory status of these active ingredients has been changing over time, and that acetochlor, dimethenamid-P, ioxynil, isoproturon and thiamethoxam are no longer approved in the European Union (European Commission, 2016). The methods of pesticide applications mostly consisted in spraying, with the exception of thiamethoxam, which was applied as seed coating. Amounts of pesticide application were not known for the year 2010 (Table 1). Pesticide amounts associated with seed coating were not known (Table 1). Pesticides were applied over a very large range of amounts, from 0.2 $g.ha^{-1}$ in the case of florasulam to 2200 $g.ha^{-1}$ in the case of isoproturon.

All of the compounds applied (Table 1) have been detected in surface waters in the course of systematic surveys of water quality carried out in Brittany from 2009 to 2015 (CORPEP, 2018). The prevalence of field-environment transfers of xenobiotics was therefore important under the prevailing climatic and agricultural conditions of the region. Moreover, these xenobiotic transfers occurred over a very wide range of chemical structures, water solubilities, Kow and DT50 dissipation half-lives (supplementary data 1; European Commission, 2016). Finally, in contrast with situations of intensive glyphosate application (Hénault-Ethier et al., 2017; Primost et al., 2017; Silva et al., 2018; Van Bruggen et al., 2018), the present context was characterized by the small contribution of glyphosate application and the important contributions of acetochlor, S-metolachlor and isoproturon applications (Table 1).

3.2. Temporal dynamics of pesticides in field and VFS soils

Annual analysis of pesticides in field and VFS soils revealed a complex pattern of persistence. As an example, analysis of VFS-114 and its adjacent field (Table 2) showed that, over 8 nominally-applied pesticides (Table 1) that were detected, two (bromoxynil, ioxynil) showed rapid dissipation or attenuation in field and VFS soils, and 5 (diflufenican, epoxiconazole, isoproturon, prochloraz, prothioconazole) showed persistence in VFS soil several months after application. Besides these persistent pesticides that could be linked to applications in the previous Spring, field and VFS soils showed the persistence of nine pesticide compounds that could not be linked to 2011 or 2010 applications, thus suggesting long-term interannual persistence over at least two years. However, some of these compounds [dimethenamid (field DT50 of 7 days), S-metolachlor (field DT50 of 23 days)] are characterized by short half-lives under laboratory or field conditions (supplementary data 1; European Commission, 2016), thus suggesting that soil contamination by pesticides may also involve landscape-scale drift or runoff between fields. Plant exposure to xenobiotics in the VFS was therefore complex, with the presence of at least 14 compounds, including 6 herbicides. Moreover, the real exposure of VFS plants to pesticides was more complex than what could be predicted from nominal applications, and therefore from global parameters of pesticide usage (Fried et al., 2018). Finally, this complexity of pesticide chemical diversity was likely to be compounded by the diversity of pesticide exposures (drift, runoff, leaching) that affect VFS plants (Gove et al., 2007; Helander et al., 2012; Rodríguez-Eugenio et al., 2018).

Quantitative analysis of pesticides in soils of VFS-91, VFS-113 and VFS-114 and in their adjacent fields (Fig. 2) confirmed that mixtures of pesticides were persistent in field and VFS soils over yearly timescales. Moreover, whereas the presence of some pesticides could be related to in-field applications of recent years (Table 1), the presence of other compounds, such as acetochlor, S-metolachlor or thiamethoxam, pointed out to landscape-scale contamination or to long-term persistence of earlier applications. Comparison of pesticide measurements in soils (Fig. 2) with expected environmental concentrations (Table 3) derived from nominal applications indicated that mitigation of applied pesticides in field soils was variable. Taking into account, as described in section 2.6, average soil characteristics that are found in northern Brittany agrosystems (Binet et al., 2006; Sulmon et al., 2007), the expected environmental concentration of epoxiconazole that was applied in February-May 2011 (Table 3) corresponds to 9.9 µg.(kg dry soil)$^{-1}$ in the field. Epoxiconazole persistence in adjacent fields of VFS-91 [6.85 µg.(kg dry soil)$^{-1}$] and VFS-113 [6.75 µg.(kg dry soil)$^{-1}$] in June 2012 (Fig. 2) was therefore at a high level that implied a DT50 > 365 days in comparison with the reported field DT50 of 98-120 days (supplementary data 1; Silva et al., 2019), thus reflecting poor mitigation of epoxiconazole in field soils. In contrast, comparison of the low residual levels of diflufenican and prochloraz (Fig. 2) with expected environmental concentrations (Table 3) indicated active mitigation of these xenobiotics in field soils. On the basis of an exponential decay described by [(Pesticide amount after 365 days)=(Pesticide initial amount)x$(2^{365/DT50})^{-1}$], the comparison of Table 3 and Fig. 2 gave DT50 estimations of 63 days for diflufenican in the field of VFS-114, in line with the reported field DT50 of 65 days (supplementary data 1), and 59 days for prochloraz in the field of VFS-114, in contrast with the reported field DT50 of 17 days (supplementary data 1).

Comparison of pesticide persistence (Fig. 2) with the year of application (Table 1), with pesticide chemical properties (supplementary data 1) and with rainfall data (supplementary data 2) did not indicate any particular trend associated with persistence or dissipation. Whereas the April-May-June rainfall of 2012 was much greater than that of 2011 (supplementary data 2), the residual pesticides that were detected (Fig. 2) had been applied both in 2011 (diflufenican, epoxiconazole, prochloraz) and in 2012 (thiamethoxam). Analysis of field soils and of the different sub-strips of VFSs (Fig. 2) showed that, in the three field-VFS systems, and for most of the contaminating compounds, there was a gradient of mitigation from the field to the distal sub-strip of the VFS. Thus, in line with numerous previous studies on VFSs and hedgerows (Collins et al., 2014; Dillaha et al., 1989; Krutz et al., 2005; Magette et al., 1989; Serra et al., 2016; Stehle et al., 2011; Thomas and Abbott, 2018), the experimental VFSs that were set up in 2010 effected significant pesticide abatement between the field and riparian compartments. Moreover, in the case of prochloraz, epoxiconazole, diflufenican, and thiamethoxam, mitigation led to the absence of detection of the xenobiotics in the distal sub-strip of the VFS, thus highlighting the role of the VFS in protecting riparian environments and streams from xenobiotic transfers. In contrast, the distinct distribution pattern of acetochlor and S-metolachlor, which were detected in sub-strip C of the VFSs, would be consistent with contamination from other sources than adjacent fields.

Comparison of the dynamics of the three triazole compounds, epoxiconazole, metconazole and prothioconazole, which were applied in the adjacent fields of the 3 VFSs (Table 3), revealed striking differences. Comparison of Fig. 2 and Table 3 showed that epoxiconazole was highly persistent, especially in the adjacent fields of VFS-91 and VFS-113. In contrast, metconazole and prothioconazole, which were applied at higher doses than epoxiconazole (Table 1), were not detected. Epoxiconazole has indeed been identified as a major and frequent contaminant of European Union soils (Silva et al., 2019). Complete dissipation of epoxiconazole in the distal sub-strip of the 3 VFSs (Fig. 2) was therefore an important feature of VFS efficiency in the present agricultural context.

3.3 Temporal dynamics of plant species within the experimental VFSs

VFSs can show a variety of designs in terms of size, positioning, or plant communities (Carluer et al., 2017; Gene et al., 2019; Hénault-Ethier et al., 2017; Serra et al., 2016). Plants with various phylogenetic, life-cycle, morphological and functional characteristics have been used (Boutin et al., 2019; Carluer et al., 2017; Cullen, 1964; Falquet et al., 2015; Gene et al., 2019; Haan et al., 1994; Hénault-Ethier et al., 2017; Serra et al., 2016; Silvertown et al., 1994; Troiani et al., 2016; Turkington and Franko, 1980). In the present study, experimental design and initial sowing of the same seed mixture at different locations resulted in identical plant communities, whose trajectories were followed over two years of agricultural practice. Botanical surveys of the VFSs (Table 4) showed striking differences between plant species with a high level of maintenance for *Festuca rubra, Lolium perenne, Agrostis stolonifera*, and *Phleum pratense* and a low level of maintenance for *Centaurea cyanus* and *Fagopyrum esculentum*. The 4 plant species that showed the highest level of maintenance were all Poaceae with a perennial lifestyle, medium to high tolerance to root-level pollutants and variable competitive ability (Table 4). The 2 plant species that showed the lowest level of maintenance were non-Poaceae with an annual lifestyle, low to medium tolerance to root-level pollutants and variable competitive ability (Table 4).

FCA of the initial botanical surveys of surface covers, carried out in June 2011, did not show any major difference of plant composition between the 5 VFSs nor between the 3 sub-strips across the 5 VFSs (data not shown). The botanical surveys that were carried out in June 2012 revealed the disappearance of *Anthemis tinctoria*, *Centaurea cyanus* and *Fagopyrum esculentum* in all of the VFSs. Moreover, FCA of these botanical surveys (Fig. 3) showed diverging trajectories between the different VFSs, with VFS-91 showing an enrichment in *Festuca rubra*, *Lotus corniculatus* and *Trifolium pratense*. This FCA (Fig. 3) also revealed the divergence of sub-strip C from the other sub-strips, with a relative increase of bare ground and the establishment of novel spontaneous vegetation in sub-strip C and an enrichment in *Lolium perenne* and *Phleum pratense* in sub-strip A. This discrimination between sub-strips A, B and C was verified for the 5 VFS and was therefore independent from the individual locations of the VFSs, thus suggesting the effects of field/non-field contrasts, that must involve not only the gradients of residual pesticides (Fig. 2), but also gradients of pesticide drifts, in-field fertilization drifts, light availability or soil properties (Boutin et al., 2019; De Snoo and Van der Poll, 1999; Guerrieri et al., 2019; Kleijn, 1996; Kleijn and Snoeijing, 1997; Kleijn and Verbeek, 2000; Silva et al., 2019).

3.4 Relationships between pesticides and plant community dynamics within the experimental VFSs

In the agricultural context of the study area and of the collaboration with local farmers, it was not practicable to set up control experimental VFSs with no-pesticide adjacent fields over the course of 3 years. The analysis of potential relationships between pesticides and plant community dynamics was therefore carried out as a cross-VFS comparison, under the assumption that the different field-VFS systems may show contrasted dynamics related to differential exposure to pesticides. Comparison of the dynamics of pesticide levels in the sub-strips (Fig. 2) and dynamics of plant surface covers (Fig. 3) was carried out by co-inertia analysis of the datasets for each VFS (VFS-91, VFS-113, VFS-114). The RV coefficient of this analysis (Robert and Escoufier, 1976) seemed to be related to the total amount of residual pesticides detected in sub-strip A of the VFS (Fig. 4). Thus, only VFS-91 was found to have a significant RV coefficient of 0.657744 with a p-value of 0.006. The corresponding distribution for VFS-91 (Fig. 5), taking into account the different transects, plant surface covers and residual pesticides, highlighted the weights of acetochlor, epoxiconazole, prochloraz and thiamethoxam in determining the dynamics of *Agrostis stolonifera*, *Lotus corniculatus*, *Trifolium pratense*, *Phleum pratense*, *Festuca rubra* and *Lolium perenne*. Acetochlor appeared to be associated with the facilitation of spontaneous vegetation. Prochloraz and epoxiconazole fungicides, as well as the insecticide thiamethoxam, appeared to be associated with the facilitation of *Phleum pratense*, *Festuca rubra* and *Lolium perenne*.

4. Discussion

4.1. Multi-level persistence of pesticidesThe different analyses of residual pesticide levels in the soils of fields and VFSs highlighted that pesticide persistence in this northern Brittany agricultural landscape was interannual (over at least two years), multi-chemical (at least 15 compounds in the soil of VFS-114 adjacent field), multi-functional (herbicides, fungicides, insecticides) and multi-source (in-field, landscape-scale). In the case of epoxiconazole, persistence in field soil, as in the adjacent field of VFS-91 [6.85 µg.(kg dry soil)$^{-1}$], represented 68% of the initial 9.9 µg.(kg dry soil)$^{-1}$ level derived from the expected environmental concentration (Peterson et al., 1994) from the application that had taken place the previous year (Table 3). Given the 98- or 120-day DT50 of epoxiconazole (supplementary data 1; Silva et al., 2019), the high level of epoxiconazole persistence after one year may have been due to specific environmental conditions within the northern Brittany landscape that remain to be characterised. Similarly, the DT50 estimation of 59 days for prochloraz (Table 3; Fig. 2), in contrast with the reported field DT50 of 17 days (supplementary data 1), suggested that local conditions led to extended persistence.

The observed multiplicity of soil pesticide contaminations, in an agricultural context of low levels of glyphosate application, was significantly higher than that reported in most European agricultural soils (Silva et al., 2019). The survey of agricultural soils that was carried out in 11 countries and 6 agricultural systems showed that detection of more than 10 pesticide residues was very rare (Silva et al., 2019). Among the 15 compounds that were detected in the present study, only two, boscalid and epoxiconazole, have been reported to be most frequently found in European soils (Silva et al., 2019). Silva et al. (2019) considered that, given the available datasets of their study, no clear conclusion could be drawn between the diversity of pesticide use and the occurrence of pesticide residues in soil. The present field-scale study highlighted the significant proportion of soil-measured pesticides (at least 15) relatively to the number of applied pesticides at field level (27). It also highlighted that the actual persistence of pesticides in field and VFS soils was more complex than what could have been predicted from nominal applications and DT50 properties (supplementary data 1; European Commission, 2016), and therefore, *a fortiori*, from global regional parameters of pesticide usage that are used in large-scale studies (Billeter et al., 2008; Fried et al., 2018; Geiger et al. 2010).

The persistence of mixtures of pesticides in the environment, even at low level, presents a potential stress risk for non-target organisms and non-target ecosystems (Alberto et al., 2016, 2017; Peterson et al., 1994; Ramel et al., 2007, 2012; Serra et al., 2013; Serra et al., 2019) and a potential health risk as possible human poisons or carcinogens (Carvalho, 2017; Krutz et al., 2005; Wilson and Tisdell, 2001). On the one hand, the VFSs were shown to function as efficient barriers to prevent xenobiotic transfers from the fields to the riparian environments. This was particularly true for epoxiconazole, which, as described above, showed a high level of persistence in field soils. However, epoxiconazole in sub-strip A of VFS-91 [4.04 µg.(kg dry soil)$^{-1}$] (Fig. 2) represented 41% of the expected environmental concentration [9.9 µg.(kg dry soil)$^{-1}$] (Peterson et al., 1994) derived from the application that had taken place the previous year (Table 3). In other words, the spread of epoxiconazole from the field was higher than most assumptions of pesticide drift (Brown et al., 2009; Gove et al., 2007; Peterson et al., 1994). It was therefore all the more striking that epoxiconazole was not detected in the distal sub-strip of the VFSs.

Mitigation efficiency of VFSs depends on VFS characteristics and processes, such as width, shielding, fungal and microbial bioremediation, or direct phytoremediation (Krutz et al., 2005; Mench et al., 2010; Roberts et al., 2012; Thomas and Abbott, 2018), as well as on physico-chemical processes linked to climate, soil, hydrology and sediment transport (Muñoz-Carpena et al., 2015, 2019). The present comparison of different VFS trajectories starting from the same initial plant community could not reveal which of these characteristics or processes played a major role in VFS-based mitigation. However, whatever the processes involved, the dynamics of the VFS plant community showed that *Festuca rubra*, *Lolium perenne*, *Agrostis stolonifera*, *Lotus corniculatus*, *Trifolium pratense* and *Phleum pratense* could successfully carry out their agri-environmental roles under the conditions of the northern Brittany landscape. On the other hand, VFS soil itself was also contaminated by S-metolachlor and acetochlor. These pesticides had not been applied in adjacent fields in the previous years, but were part of pesticide usage in the area, as exemplified by their use in the adjacent fields of VFS-109 and VFS-112 (Table 1), thus indicating the existence of landscape-scale processes of dissemination (Collins et al., 2014). Imidacloprid, alachlor, boscalid, bromuconazole and cadusafos were also detected in both field and VFS soils (Table 2), although they had not been used in any of the fields under study, thus suggesting that landscape-scale contamination could occur over long distances.

4.2. Effects of pesticide dissemination and soil persistence on plant community dynamics

Our present field-scale approach, combined with actual measurements of pesticides in soils, revealed contrasted situations between fields and between VFSs within the same agricultural landscape, thus highlighting the difficulty to correlate plant dynamics and global levels of pesticide usage. Moreover, even at field level, these contrasts (Fig. 2) did not directly depend on the nominal amount of pesticide applications (Table 1). Similar levels of application resulted in different dynamics of pesticides in the soils, which was also compounded by contamination by landscape-level pesticides. These contrasts had differential effects on plant dynamics in the different VFSs under study, with one particular VFS only, VFS-91, showing a significant correlation between plant dynamics and pesticide levels in the soil (Figs 4-5). Conversely, the absence of correlation in VFS-113 and VFS-114 highlighted that residual pesticides were not necessarily the major driving factor of plant dynamics in all of the VFSs and that other factors or parameters were involved, either as direct drivers of plant dynamics or in interaction with residual pesticides. Additional multi-level studies would therefore be required to take into account physico-chemical factors such as soil and hydrological properties, as well as the modalities of pesticide transport from the field to accumulation in VFS soil.

It could be tentatively hypothesised that correlation between plant dynamics and residual pesticides depended on a threshold of pesticide accumulation in the soil (Fig. 4). It was noteworthy that this apparent threshold (between 2.697 and 4.772 $\mu g.kg^{-1}$ dry soil) that could be derived from the 12-pesticide screening given in Fig. 2 was lower than the sums of the pesticide levels (adjacent field of VFS-91: 10.561 $\mu g.kg^{-1}$ dry soil; adjacent field of VFS-113: 9.471 $\mu g.kg^{-1}$ dry soil; adjacent field of VFS-114: 4.842 $\mu g.kg^{-1}$ dry soil) measured in the adjacent fields of the three locations under study and derived from the same 12-pesticide screening (Fig. 2). Changes of plant community dynamics are necessarily related to modifications of plant physiology, whether through direct effects on plant functioning or through indirect effects on plant microbial, fungal or animal environments. The levels of mixtures of residual pesticides in field soils and their interannual persistence therefore pointed out to potential carry-over effects on crops and weeds within the fields from one growing season to the other. Such effects are likely to lead to crop injury, weed control failure or induction of herbicide resistance (Alberto et al., 2016; Serra et al., 2019). Moreover, these carry-over effects may hamper the transition of field use to organic farming or to specialty crops. The worldwide worry of the Food and Agriculture Organization concerning soil pollution and related yield decreases (Rodríguez-Eugenio et al., 2018) should therefore not be ruled out at the local and regional European scale.

The plant-pesticide relationship that was found in VFS-91 (Fig. 5) emphasised negative correlation of the herbicide acetochlor with *Trifolium pratense*, and positive correlations of the insecticide thiamethoxam and of the fungicides prochloraz and epoxiconazole with

Festuca rubra, *Lolium perenne* and *Phleum pratense*. Studies under axenic, *in vitro* or artificial conditions show that plants can uptake xenobiotics at root level with subsequent root-shoot translocation (Mörtl et al., 2019; Sulmon et al., 2007) and that low, environmentally-relevant, sublethal and subtoxic levels of not only herbicides, but also fungicides and insecticides, can directly affect plant metabolism and physiology (Alberto et al., 2017, 2018; Saunders et al., 2013; Serra et al., 2013, 2015a, 2015b, 2019; Soares et al., 2019).

Acetochlor, triazoles, prochloraz and thiamethoxam can indeed directly interact with plant growth and development. The acetochlor herbicide has a direct effect on protein synthesis (Mezzari et al., 2005). The fungicide tebuconazole, which belongs to the same triazole family as epoxiconazole, and which is a major contaminant of European soils (Silva et al., 2019), has negative effects on root growth of *Anthemis tinctoria* and *Centaurea cyanus* and positive effects on root growth of *Lolium perenne* (Serra et al., 2019). The fungicide prochloraz has been shown to interfere with plant brassinosteroid and cytokinin regulations (Werbrouck and Debergh, 1996, 2004). Thiamethoxam has been shown to interfere with plant drought stress responses and salicylate regulations (Ford et al., 2010; Stamm et al., 2014).

The disappearance of *Anthemis tinctoria* and *Centaurea cyanus* from all of the VFSs could be related to their sensitivity to agricultural pollutants (Serra et al., 2019), and conversely, *Festuca rubra*, *Lolium perenne*, *Agrostis stolonifera*, *Lotus corniculatus* and *Trifolium pratense*, which were maintained in the VFSs, show positive growth responses to some agricultural pollutants (Serra et al., 2019). These relationships could thus reflect direct effects of pesticides on plant dynamics in the agricultural landscape under study. In contrast, the disappearance of *Fagopyrum esculentum* from all of the VFSs could not be ascribed to a higher sensitivity to xenobiotics, and the maintenance of *Phleum pratense* in the VFS contrasted with its negative growth responses to agricultural pollutants (Serra et al., 2019).

Further work is therefore required to analyse the *in situ* responses of plants to contaminating pesticides, in terms of uptake and physiological perturbations, and to clarify the interferences between the responses to contaminating pesticides and other influencing factors, such as plant interspecific competition (Boutin et al., 2019; Damgaard et al., 2014; Jung et al., 2009), fertilizers (Gove et al., 2007; Pellissier et al., 2014) or soil and hydrological properties (Gove et al., 2007; Muñoz-Carpena et al., 2015, 2019).

4.3. Potential consequences for environmental risk assessment

Given their potential impact on plants and ecosystem services, detection of residual pesticides (herbicides, fungicides, insecticides) may be construed as a situation of agricultural soil pollution (Rodríguez-Eugenio et al., 2018). The importance of acetochlor, epoxiconazole, prochloraz and thiamethoxam in terms of persistence and potential impacts must be modulated in view of the constant evolution of pesticide regulations. In the European Union (European Commission, 2016), whereas prochloraz is still an approved pesticide, thiamethoxam and acetochlor are no longer approved, and epoxiconazole is in a process of expiration of approval. Moreover, acetochlor, epoxiconazole, prochloraz and thiamethoxam remain in use in other countries, such as Australia (https://portal.apvma.gov.au).

The relative importance of the fungicides epoxiconazole and prochloraz in terms of persistence in soils, residual levels and impact on vegetation dynamics underlined the need to give more attention to environmental risks associated with fungicides (Satapute et al., 2019; Zubrod et al., 2019). Epoxiconazole is detected in surface waters at a mean concentration of 2 (± 8, SEM) $\mu g.L^{-1}$ (Zubrod et al., 2019). Epoxiconazole concentrations measured in the fields and VFS of the present study were therefore within the higher values of that range. Moreover, both triazoles tebuconazole and epoxiconazole have been reported to be highly toxic to non-target fungi, such as aquatic fungi, with significant effects at 1 $\mu g.L^{-1}$ (Dijksterhuis et al., 2011). Tebuconazole has also been reported to inhibit metabolism of microbial communities at a concentration of 2 µg.L^{-1} (Artigas et al., 2014). The direct negative effects of tebuconazole on plants have been shown to occur at much higher concentrations in the range 300-1200 µg.L^{-1} (Serra et al., 2013, 2019), which would suggest that the effects on vegetation dynamics described above could not be ascribed to direct effects of triazoles on plants. However, direct effects due to the combination of residual pesticides (acetochlor, epoxiconazole, prochloraz, thiamethoxam) could not be ruled out. At any rate, the present study strongly indicated that residual levels of triazoles were highly likely to affect microbial communities and generate microbial perturbations that can affect soil health and plant growth and development (Satapute et al., 2019; Zubrod et al., 2019).

Besides their central roles in pesticide and nutrient mitigation, VFS can be designed to support other ecosystem services (Gene et al., 2019; Hille et al., 2018). Direct or indirect effects of residual pesticides leading to modifications of VFS vegetation may not be innocuous in terms of maintenance of such ecosystem services. In a slightly different context, that of field margins, Pollier et al. (2018) have emphasised the relationships between vegetation and arthropod dynamics and advocated the protection of field margins from pesticide drift, runoff and leaching. Yamamuro et al. (2019) have demonstrated the long-term and cascading effects of pesticide impacts in the environment. Moreover, low levels of residual pesticides can affect plant metabolism of soluble sugars, organic acids and amino acids without necessarily affecting plant survival and growth (Serra et al., 2013), and therefore induce cascading effects on feeding stimulation of insects (Hervé et al., 2014) or nitrogen quality of forage for livestock and wild animals (Savary-Auzeloux et al., 2003).

The levels of epoxiconazole persistence in field soils (Fig. 2) would theoretically entail the presence of 23 g of epoxiconazole at the level of a one-ha field, such as the adjacent field of VFS-91. The levels of epoxiconazole persistence in sub-strip A (Fig. 2) would correspond to the presence of 13.5 g of epoxiconazole in the theoretical context of two 2m wide VFS ribbons on either side of a 5000-m stream. Climate-related events, through flooding from the streams or through wind-borne dust from fields and VFSs, and agricultural mechanical work, through dust-forming processes or through road traffic of agricultural machinery, are likely to spread fractions of these levels of persistent xenobiotics throughout the landscape in a manner that bypasses VFS barriers. Thus, flood events with pesticide discharges of 0.34 $g.ha^{-1}$ have been shown to lead to stream contaminations above maximum allowable concentrations (Rabiet et al., 2010). All of this strongly suggested that xenobiotic mitigation must include landscape-scale practices, aiming in particular at the control of field erosion, sediment transport and runoff processes (Muñoz-Carpena et al., 2015, 2019; Gene et al., 2019; Silva et al., 2019), in order to complement VFS-based processes.

4.4. Potential consequences for animal and human health

Acetochlor, epoxiconazole and thiamethoxam are considered to be cytotoxic, hepatotoxic and/or carcinogenic (Green et al., 2005; Heise et al., 2018; Kale et al., 2008), and additive or potentiating toxicological effects of mixtures of herbicides, fungicides and insecticides have been described (Reffstrup et al., 2010; Rizzati et al., 2016). The long-term persistence of pesticides in agricultural and VFS soils in the landscape under study may therefore imply that soil and dust contacts can have negative effects on animal wildlife and livestock (Bart et al., 2019; Van Bruggen et al., 2018; Wilson and Tisdell, 2001). It also implies that agricultural activities take place in a contaminated environment, with therefore potential impact on agro-workers and agro-users.

The European Food Safety Authority (EFSA) considers that there are many unknowns regarding the pathways of non-dietary exposure to pesticides (EFSA, 2014). It has been established that dermal exposure and inhalation are important features of occupational hazards linked to pesticide preparation and application (Flack et al., 2008). Moreover, novel scenarios of exposure, such as contact with pesticide-contaminated waters, are increasingly taken into account (Bányiová et al., 2019). Thus, in a situation of contaminated agricultural and VFS soils, as described above, dust-forming processes in Summer and Autumn or mud-forming conditions from Autumn to Spring may result in inhalation of or skin exposure to pesticide-laden particles, especially affecting operators and workers. On the other hand, acceptable daily intakes or reference doses are highly variable between pesticides, ranging from $\mu g.kg^{-1}.day^{-1}$ to $mg.kg^{-1}.day^{-1}$ orders of magnitude (Bányiová et al., 2019). Further research is therefore required to diversify health risk assessments in pesticide-contaminated environments (Bányiová et al., 2019) within a framework of ongoing multi-level vigilance that should supersede the current system of regulatory approval (Milner and Boyd, 2017).

Conflict of interest

The authors do not have any commercial or financial conflict of interest regarding the present article.

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Legends of Figures

Figure 1. Study area and lay-out of the field/VFS/riparian environment continuum. The location (48° 36' N, 1° 32' W, Pleine-Fougères, Ille-et-Vilaine, Brittany, France) of the Zone Atelier Armorique Long-Term Ecological Research (LTER) network (red star) is positioned on the maps of Brittany and Europe. For each of the five VFSs under study, proximal, central and distal sub-strips relatively to the adjacent cultivated field were respectively identified as A, B and C. In each VFS, sampling of soils and vegetation surveys were carried out along five perpendicular transects with sampling points in the adjacent field and the three sub-strips (red, black, blue, green, purple spots), thus leading to 5 sampling or observation points per sub-strip (A1, A2, A3, A4, A5/ B1, B2, B3, B4, B5/ C1, C2, C3, C4, C5). The five perpendicular transects were parallel to each other and at 20 m from each other.

Figure 2. Residual levels of pesticides in the soils of VFSs and of their adjacent fields. A set of 12 pesticides (diflufenican, dimethenamid-P, S-metolachlor, acetochlor, pendimethalin, prochloraz, thiamethoxam, flufenacet, cypermethrin, epoxiconazole, metconazole, prothioconazole) that were among the 28 pesticides nominally applied to the fields adjacent to the different VFSs under study (Table 1) was analysed by GC-MS in soils sampled in June 2012. Six pesticides (prochloraz, epoxiconazole, diflufenican, thiamethoxam, S-metolachlor, acetochlor) were detected and quantified in the VFSs under study (VFS-91, VFS-113, VFS-114) and their adjacent fields. Statistical significance of differences ($P \leq 0.05$) is shown by asterisks or lower-case letters near the bars.

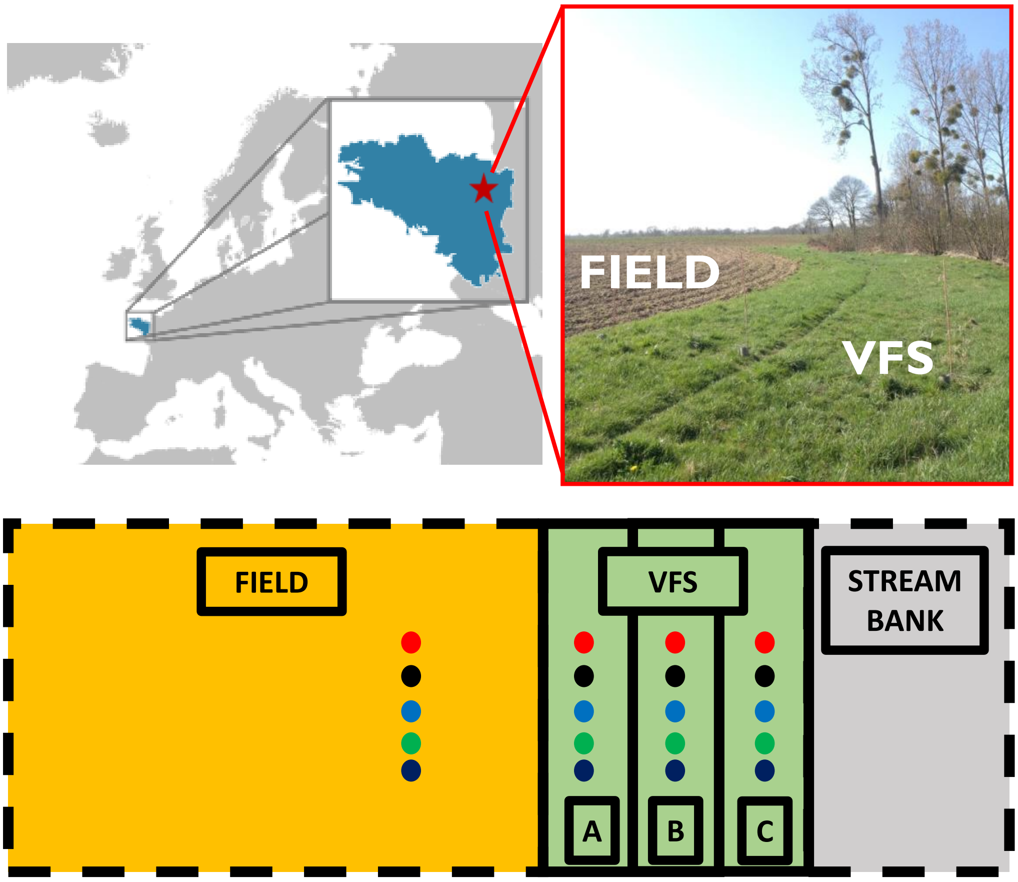
Figure 3. Correspondence factor analysis of plant-species-specific surface cover according to VFS and according to VFS sub-strip. Identification of plant species and quantification of plant-species-specific, additional spontaneous vegetation and bare ground percentages were carried out in June 2012. Plant species from the initial sowing are described by their generic name. Additional plant species that were found in the VFSs are described as other species.

FCA was carried out on the correlation matrix of plant species covers that were determined in the various VFSs and sub-strips (Fig. 1). The most informative two-dimensional representation (dimension 1, dimension 2) of the data is shown, with the two factorial axes accounting for 21.8% and 19.5% of the total inertia of the data analysed (inset). (A) Distribution of plant species, (B) Correlated distribution of the VFSs under study (VFS-91, VFS-109, VFS-112, VFS-113, VFS-114) relatively to plant species covers, (C) Correlated distribution of the sub-strips (Fig. 1) relatively to plant species covers. Colors highlight the different VFSs (B) or the different sub-strips (C). Individual determinations and their distance to the position of the weighted average are shown as dots and solid lines of the same color as their respective VFS (B) or sub-strip (C).

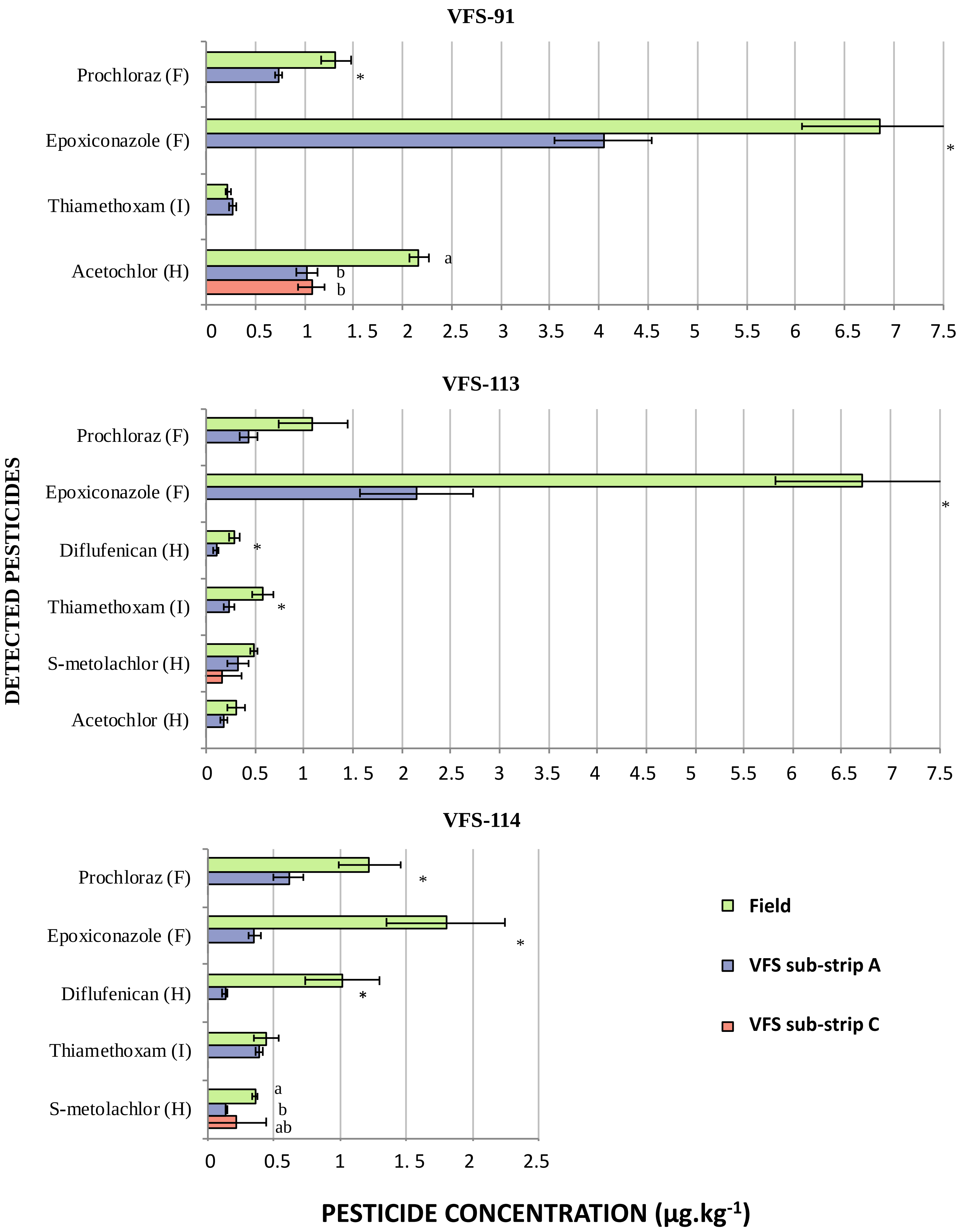
Figure 4. Relationship between RV coefficients of co-inertia analysis and residual soil levels of total pesticides in the VFSs. Co-inertia analysis was carried out on the correlations between residual soil levels of individual pesticides in sub-strip A (Fig. 2) and plant-species-specific, additional spontaneous vegetation, and bare ground surface cover percentages in the VFS (Fig. 3). The resulting RV coefficients were plotted against the sum of residual pesticide levels measured in sub-strip A of VFS-91, VFS-113 and VFS-114 (Fig. 2).

Figure 5. Co-inertia analysis between plant-species-specific surface covers and residual soil levels of individual pesticides in VFS-91. The co-inertia analysis that was carried out on the correlations between residual soil levels of individual pesticides (Fig. 2) and plant-species-specific, additional spontaneous vegetation, and bare ground surface covers (Fig. 3) in the VFSs, gave a significant RV coefficient only in the case of VFS-91 (Fig. 4). The correlation of distribution between residual pesticides and plant-species-specific surface covers was therefore further analysed in the 5 transects of the different sub-strips of VFS-91 (Fig. 1). Plant species under study are described by their generic name. (A) Distribution of the sampling points in sub-strip A and sub-strip C of VFS-91 [A1-A5, circled in blue: sampling

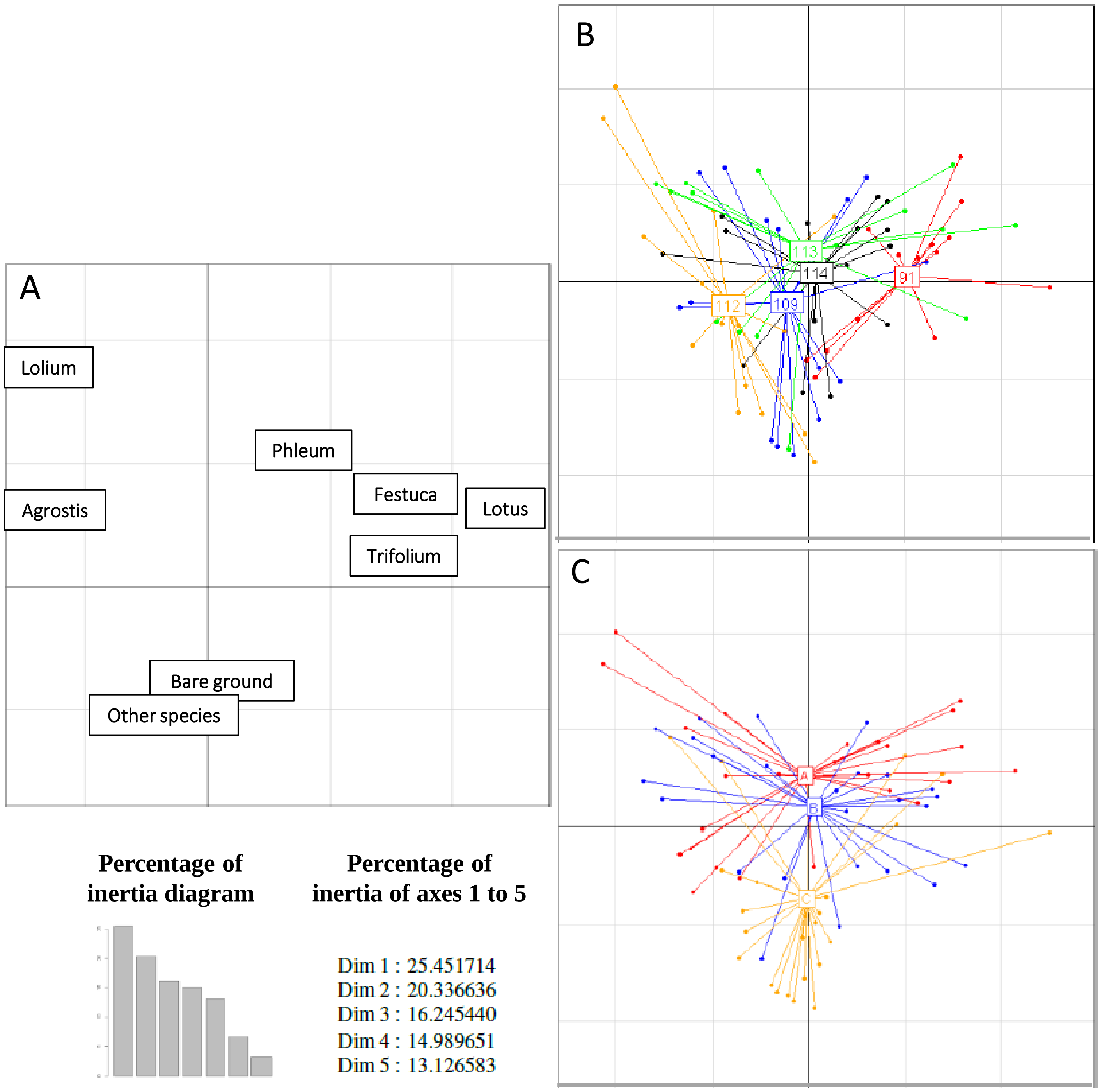
points for the 5 transects of sub-strip A (Fig. 1); C1-C5, circled in green: samplings points for the 5 transects of sub-strip C (Fig. 1)], (B) Position of the parameters of plant-species-specific covers in VFS-91, (C) Position of residual pesticide parameters in VFS-91. Arrows (A) represent co-inertia axes of the relationship between plant cover patterns (base of the arrow) and residual pesticide levels (arrowhead). The two lower panels are the projection of plant surface cover (B) and pesticide (C) characteristics onto the co-inertia plane.



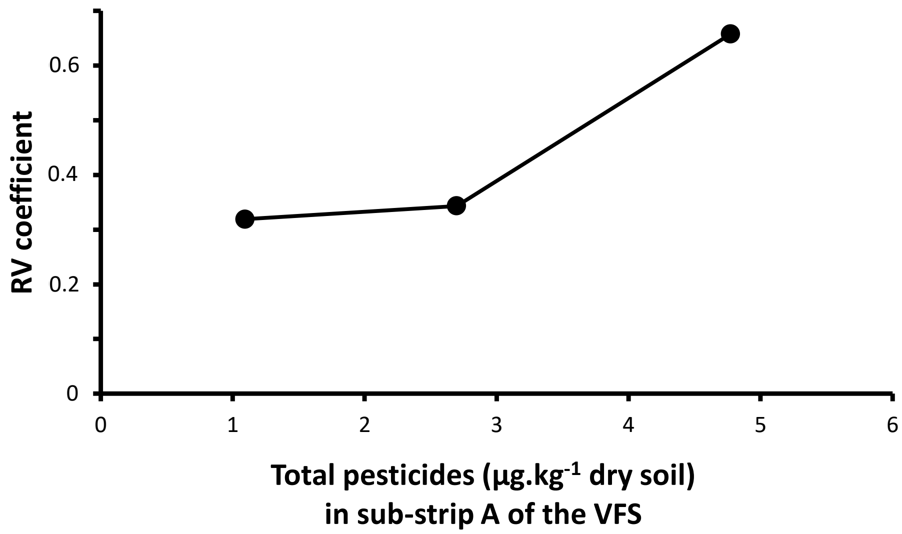
FIELD
VFS
FIELD
VFS
STREAM BANK
A
B
C



VFS-91
Prochloraz (F)
Epoxiconazole (F)
Thiamethoxam (I)
Acetochlor (H)
a
b
b
*
*
0
0.5
1
1. 5
2
2.5
3
3.5
4
4.5
5
5.5
6
6.5
7
7.5
VFS-113
Prochloraz (F)
Epoxiconazole (F)
Diflufenican (H)
Thiamethoxam (I)
S-metolachlor (H)
Acetochlor (H)
*
*
*
VFS-114
Prochloraz (F)
Epoxiconazole (F)
Diflufenican (H)
Thiamethoxam (I)
S-metolachlor (H)
a
b
ab
*
*
*
0
0.5
1
1. 5
2
2.5
Field
VFS sub-strip A
VFS sub-strip C
DETECTED PESTICIDES
PESTICIDE CONCENTRATION (µg.kg-1)



A
Lolium
Phleum
Agrostis
Festuca
Lotus
Trifolium
Bare ground
Other species
B
113
114
91
112
109
C
A
B
C
Percentage of inertia diagram
Percentage of inertia of axes 1 to 5
Dim 1 : 25.451714
Dim 2 : 20.336636
Dim 3 : 16.245440
Dim 4 : 14.989651
Dim 5 : 13.126583


RV coefficient
0.6
0.4
0.2
0
0
1
2
3
4
5
6
Total pesticides (µg.kg-1 dry soil)
in sub-strip A of the VFS

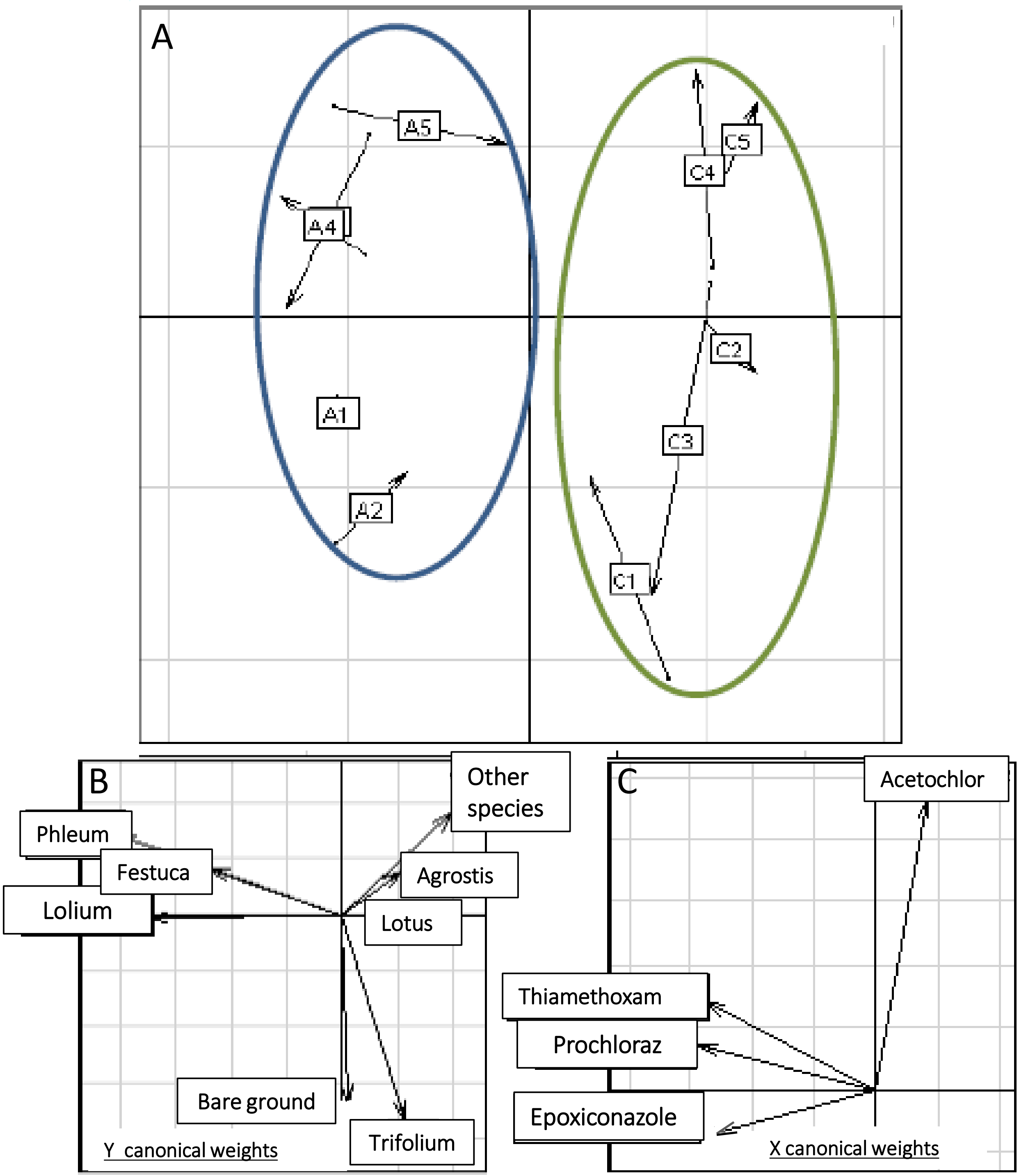

A
A5
A4
A1
A2
C5
C4
C2
C3
C1
B
Other species
Phleum
Festuca
Agrostis
Lolium
Lotus
Bare ground
Trifolium
Y canonical weights
C
Acetochlor
Thiamethoxam
Prochloraz
Epoxiconazole
X canonical weights

Table 1. Amounts of pesticide active ingredient applications (g/ha) in adjacent fields of the VFSs over the 2010-2012 period. Data of pesticide commercial product applications were obtained from the farmers in charge of the different fields, and the levels of active ingredient applications were derived from commercially-available characteristics of the products. Data for the year 2010 consisted in the nature of applied active ingredients with no information on the amounts involved. Pesticide treatments were carried out between February and May. F: fungicide; H: herbicide; I: insecticide; NA: amount of application not available; SC: unknown amount of seed-coating-associated pesticide.

	Year 2010					Year 2011					Year 2012				
	VFS 91	VFS 109	VFS 112	VFS 113	VFS 114	VFS 91	VFS 109	VFS 112	VFS 113	VFS 114	VFS 91	VFS 109	VFS 112	VFS 113	VFS 114
Dicamba (H)			NA					480			480	150			
Diflufenican (H)									178	178					
Dimethenamid-P (H)	NA	NA										318.8			
S-metolachlor (H)			NA					1000					1200		
Acetochlor (H)							1101					367			
Fluroxypyr (H)												20			
Pendimethalin (H)	NA						800					375	400		
Bromoxynil (H)		NA		NA	NA		135		127.4	127.4	405	60		337.5	337.5
Ioxynil (H)									70.4	70.4					
Prochloraz (F)						315			315	315					
Thiamethoxam (I)								SC				SC		SC	SC
Glyphosate (H)												12.3			
Cypermethrin (I)							80								
Trifloxystrobin (F)						350			350	350					
Nicosulfuron (H)	NA		NA	NA	NA			12			28	10	12	12	12
Prosulfuron (H)			NA					7.5			7.5	3	7.5		
Iodosulfuron-methyl Na+ (H)						4.5									
Mesosulfuron-methyl Na+ (H)						4.5									
Tribenuron-methyl (H)						3.6									
Metsulfuron-methyl (H)						3.6									
Epoxiconazole (F)						33.3			33.3	33.3					
Metconazole (F)						72			72	72					
Prothioconazole (F)						175			175	175					

Florasulam (H)														0.2			
Sulcotrione (H)	NA	NA	NA														
Mesotrione (H)				NA	NA					100			110	60	120	30	30
Isoproturon (H)										2200	2200						
Flufenacet (H)														408			
Total pesticides	NA	NA	NA	NA	NA		961.5	2116	1599.5	3521	3521		1030.5	1376	1739	379.5	379.5

Table 2. Annual dynamics of pesticide detection in VFS-114 and its adjacent field. A set of 22 pesticides (fungicides, herbicides, insecticides) was analysed by LC-MS/MS (Rovaltain Research Company, Alixan, France) in soil samples from the field and sub-strip A of VFS-114. Quantifications were carried out on determinations from 3 transect samples taken at random from the 5 transect samples (Fig. 1). F: fungicide; H: herbicide; I: insecticide; X: detected; ND: not detected

Pesticide	Field application February -May 2011	Detection in field soil June 2011	Detection in VFS soil June 2011	Detection in VFS soil October 2011
Bromoxynil (H)	Yes	ND	ND	ND
Ioxynil (H)	Yes	ND	ND	ND
Metconazole (F)	Yes	X	ND	ND
Diflufenican (H)	Yes	X	X	X
Epoxiconazole (F)	Yes	X	X	X
Isoproturon (H)	Yes	X	X	X
Prochloraz (F)	Yes	X	X	X
Prothioconazole (F)	Yes	X	X	X
Acetochlor (H)	No	ND	ND	ND
Cypermethrin (I)	No	ND	ND	ND
Dicamba (H)	No	ND	ND	ND
Flufenacet (H)	No	ND	ND	ND
Fluroxypyr (H)	No	ND	ND	ND
Dimethenamid-P (H)	No	X	X	X
Pendimethalin (H)	No	X	X	X
S-metolachlor (H)	No	X	X	X
Thiamethoxam (I)	No	X	X	X
Imidacloprid (I)	No	X	X	X
Alachlor (H)	No	X	X	X
Boscalid (I)	No	X	X	X
Bromuconazole (F)	No	X	X	X
Cadusafos (I)	No	X	X	X

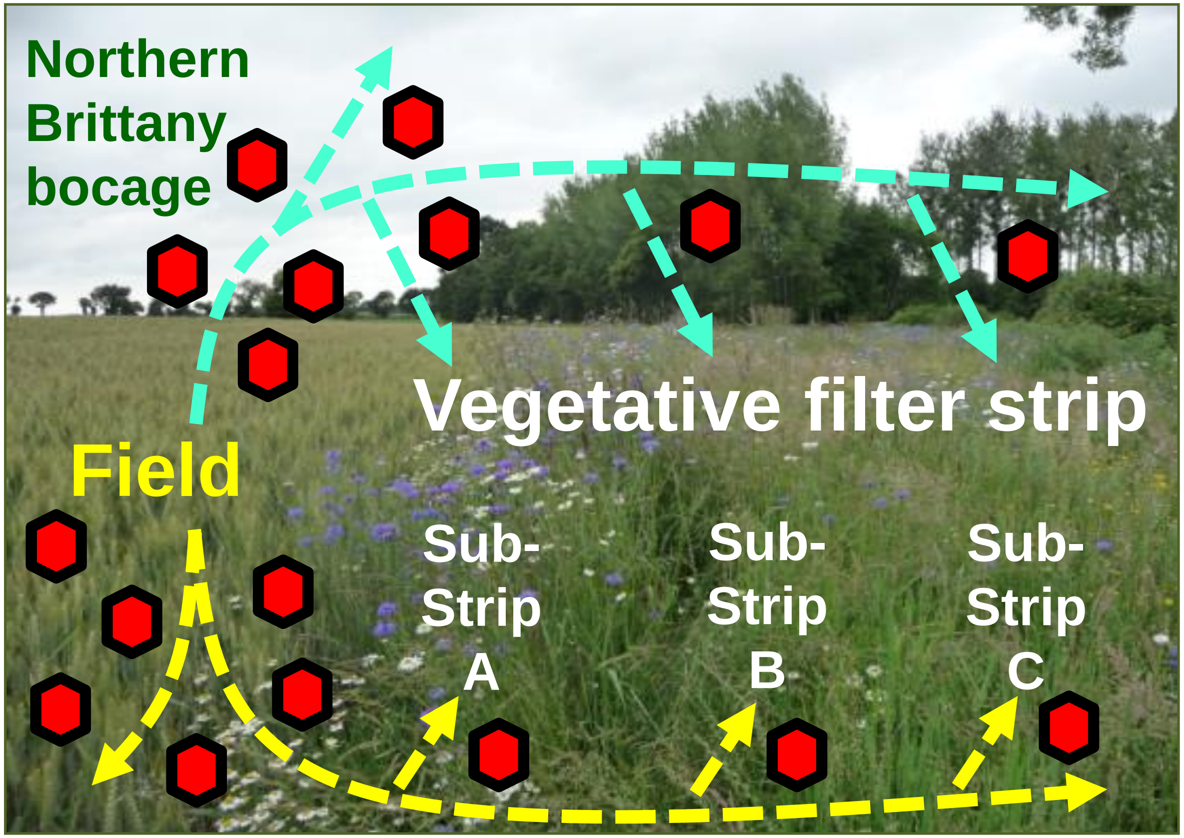
Table 3. Expected environmental concentrations of pesticide applications in adjacent fields of VFSs. The set of 12 pesticides analysed in Fig. 2 was taken into account. Expected environmental concentrations resulting from the most recent application (indicated in brackets) prior to pesticide quantification in soils in 2012 (Figure 2) were calculated as previously described (Peterson et al., 1994) on the basis of active ingredient applications (Table 1) and of a 30-cm depth of potentially affected soil. These estimations in µg per L fresh soil were converted into µg per kg dry soil estimations by division with a bulk density of 1.4 kg.L-1 and an 80% dry soil content [soil moisture: 20% (w/w)]. Pesticide applications were carried out between February and May in 2010, 2011 or 2012 (indicated in brackets). Pesticides that were actually detected and quantified in field and VFS soils (Figure 2) are underlined and in upper case. 0: no recorded application in 2010, 2011 and 2012; NA: amount of application not available; SC: unknown amount of seed-coating-associated pesticide**.**

	Expected environmental concentration (µg per kg dry soil)		
Pesticide	VFS-91 adjacent field	VFS-113 adjacent field	VFS-114 adjacent field
PROCHLORAZ	93.75 (2011)	93.75 (2011)	93.75 (2011)
EPOXICONAZOLE	9.91 (2011)	9.91 (2011)	9.91 (2011)
DIFLUFENICAN	0	52.95 (2011)	52.95 (2011)
THIAMETHOXAM	0	SC (2012)	SC (2012)
Dimethenamid	NA (2010)	0	0
Pendimethalin	NA (2010)	0	0
Flufenacet	0	0	0
Cypermethrin	0	0	0
Metconazole	21.42 (2011)	21.42 (2011)	21.42 (2011)
Prothioconazole	52.05 (2011)	52.05 (2011)	52.05 (2011)

S-METOLACHLOR	0	0	0
ACETOCHLOR	0	0	0

Table 4. Sustainability of plant species establishment in VFSs. Initial sowing of plant species in experimental VFSs was carried out in 2010. Identification of plants was carried out in the five different VFSs (VFS-91, VFS-109, VFS-112, VFS-113, VFS-114) in June 2011, October 2011, March 2012 and June 2012. The frequency of identification corresponds to the ratio of identifications over the 20 botanical surveys that were carried out. Cross-pollutant (pesticides, polycyclic aromatic hydrocarbons, heavy metals) tolerance is as described in Serra et al. (2019). Competitive ability is given as a tentative level (high, medium or low) derived from the literature (Boutin et al., 2019; Cullen, 1964; Falquet et al., 2015; Silvertown et al., 1994; Troiani et al., 2016; Turkington and Franko, 1980).

Plant species	Frequency of identification in VFSs	Family	Pollutant tolerance at root level	Competitive ability	Life cycle
Festuca rubra	1	Poaceae	8/8	Medium-High	Perennial
Lolium perenne	1	Poaceae	8/8	High	Perennial
Agrostis stolonifera	1	Poaceae	6/8	High	Perennial
Phleum pratense	1	Poaceae	5/8	Low	Perennial
Trifolium pratense	0.9	Fabaceae	5/8	Medium	Annual
Lotus corniculatus	0.7	Fabaceae	5/8	Medium	Perennial
Anthemis tinctoria	0.45	Asteraceae	0/8	Low	Perennial
Centaurea cyanus	0.3	Asteraceae	2/8	Medium	Annual
Fagopyrum esculentum	0.1	Polygonaceae	5/8	High	Annual


Northern
Brittany
bocage
Vegetative filter strip
Field
Sub-
Strip
A
Sub-
Strip
B
Sub-
Strip
C