



## Research article

# Concentration levels of new-generation fungicides in throughfall released by foliar wash-off from vineyards



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## ABSTRACT

The presence of agricultural pesticides in the environment and their effects on ecosystems are major concerns addressed in a significant number of articles. However, limited information is available on the pesticide concentrations released from crops. This study reports losses of new-generation fungicides by foliar wash-off from vineyards and their potential impact on the concentrations of their main active substances (AS) in surface waters.

Two experimental plots devoted to vineyards were treated with various combinations of commercial new-generation fungicide formulations. Then, up to sixteen throughfall collectors were installed under the canopy. Concentrations of sixteen different AS in throughfall were determined along nine rainfall episodes. Concentrations in throughfall far exceeded the maximum permissible levels for drinking water established by the European Union regulations. Dynamics of fungicide release indicated a first-flush effect in the wash-off founding the highest concentrations of AS in the first rain episodes after application of the fungicides.

This article shows that foliar spray application of commercial formulations of new-generation fungicides does not prevent the release of their AS to soil or the runoff. Concentration data obtained in this research can be valuable in supporting the assessment of environmental effects of new-generation fungicides and modeling their environmental fate.

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## 1. Introduction

The presence of pesticides in water, soils and sediments has detrimental effects on health, aquatic life, microbial communities, and invertebrates (Carvalho, 2017 and references therein). Pesticides, and more specifically, fungicides cause disruptive effects on organisms and interferences on organic matter and nutrients cycles (Hussain et al., 2009; Imfeld and Vuilleumier, 2012; Pelosi et al., 2014). Important issues related to their fate in the environment are mechanisms of controlling the fungicide release from treated crops and their transport to several environmental compartments (Cohen et al., 1995; Wauchope, 1978). Depth fungicide penetration into soil may occur mostly by preferential transport because these are subject to significant adsorption and decay (Flury, 1996). Despite the efforts on the development of quantitative models for

describing pesticide transformation pathways in the environment such as biodegradation (Hussain et al., 2015), sorption and bio-absorption (Khuntong et al., 2010; McLaughlin et al., 2016), lixiviation (Bedmar et al., 2013), or runoff in soil and water compartments (Arias-Estévez et al., 2008; Herrero-Hernández et al., 2016; Pateiro-Moure et al., 2013; Pierlot et al., 2017; Tadeo, 2008), little is known about the actual fluxes which transport the pesticides to the environment, especially in agroecosystems, and their controlling mechanisms, in particular during rainfall. For instance, pesticide removal by foliar wash-off and transport via throughfall out of the crop are the primary mechanisms involved in the release of pesticides to the environment (Cohen and Steinmetz, 1986). Throughfall may infiltrate into the soil where AS can be absorbed by soil constituents or metabolized by biota or transported over long distances on the surface runoff (Schulz, 2001; Wauchope et al., 2004). Another transport pathway is the facilitated transport of pesticides attached to soil colloids as suspended load in runoff (Wu et al., 2004).

There are some studies about foliar wash-off induced losses of

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well-known second and third generation pesticides (Fife and Nokes, 2002; Hunsche et al., 2007, 2011; Otero et al., 2011). However, continuous development of new AS and new commercial formulations requires updated investigations about their behavior in the environment.

Agricultural use of new generation fungicides, also as so-called fourth generation fungicides, is now widespread (Köller, 1999; Paranjape et al., 2014). The current development of new fungicides is focused in two main ways, namely, specificity in control the target organisms and to overcome the acquired resistance against old active substances. The environmental advantages of these strategies are reducing disruption of metabolic pathways in the non-target organisms and reducing the dose (Gisi and Sierotzki, 2008; Köller et al., 2002; Thind, 2011). However, to date, there are not scientific studies reporting their losses by foliar wash-off under natural rainfall.

The objective of this study is to quantify the concentrations of new-generation fungicides in throughfall waters from vineyards cultivated under conventional management along with the cycle crop. Knowing the average and peak concentrations along natural rainfall episodes may contribute to improving the accuracy of the estimation of the fluxes of these agrochemicals in the environment.

## 2. Material and methods

### 2.1. Study area

The study area comprises two adjacent plots devoted to vines (V1 and V2) located in the province of Ourense in the NW of Spain 42°18'02"N 8°07'53"W (Fig. 1). Mean annual temperature is 14.5 °C, 1915 h of insolation/year and 950 mm/year of total rainfall. V1 and V2 covered an area surface of 1600 and 1000 m<sup>2</sup> respectively to produce white grapes of *Vitis vinifera* var. Godello which is a local variety. The vines are 1 m distanced from each other within a row and 1.8 m between the rows. The plants were grown using a vertical trellis system with a canopy on average of 80 cm width at their maximum development. The vineyard plots were monitored for more than a year, from 21/08/2008 till 18/11/2009 which comprising two natural years as well as two grape harvests (19/09/2008 and 21/09/2009).

### 2.2. Fungicide applications

The application strategy followed the current schedule used by farmers using doses recommended by the manufacturer. (Sub)plots with different treatments were designed to cover a wide range of

commercial formulations combinations. Thirteen commercial formulations of fungicides (see S1 of Supplementary information) were applied in up to two vineyards following various phases according to Table 1, in various randomized (sub)plots (Fig. 2). A total of ten successive applications (A1–A10) were sprayed on vines to control downy mildew, powdery mildew and grey mould. Applications were grouped into three phases: phase i) the first application (A1) was carried out using two types of formulations grouped taking into account the target pest: on one hand, five commercial formulations were sprayed to fight against downy mildew, each within their respective subplots (A–F), namely Cabrio Top (A), Equation Pro (B), Fantic M (C), Mikal Premium (D), one control (E) (without treatment against downy mildew) and Fobeci (F) (Fig. 2). On the other hand, the other types of formulations used in A1 consisted of spraying Topas EC10 and Switch in the whole vineyard V1 to control powdery mildew and grey mould, respectively, following the schedule displayed in Table 1 and Fig. 2. Phase ii) which consisted of spraying Cabrio Top (A2 and A3) and Fobeci (A4) in both whole vineyards V1 and V2 just to control downy mildew (no applications were carried out to control neither powdery mildew nor grey mould) (Fig. 2).

In phase iii) applications from 5 to 10 were conducted applying four different treatments to control downy mildew (mandipropamid, valifenalate, Mildicut and Equation Pro), two different treatments for powdery mildew (Talendo and Vivando) and two separate treatments to control grey mould (Cantus and Switch) as it is shown in Fig. 2. It should be noted that mandipropamid and valifenalate were applied as AS due to they were pending of phytosanitary registration in Spain in that moment. All applications were performed using an M-83-E hand-gun sprayer (Sirfran, Alicante, Spain) ensuring that bunches and leaves were well covered. The fungicide treatment schedule is the same as described in González-Rodríguez et al. (2011) and all dates of application are also shown in Fig. 2. The volume of the application depended on the phenological stage: 60 L ha<sup>-1</sup> for the first application, 120 L ha<sup>-1</sup> for second and third applications and 240 L ha<sup>-1</sup> for subsequent applications.

### 2.3. Rainfall records

Rainfall was recorded throughout the experiment using a tipping-bucket pluviometer (0.2 mm per stroke) every ten minutes. Rainfall episodes (RE) were recorded separately using the criterion that an individual episode is that is separated by at least one day with less than 1 mm rainfall.

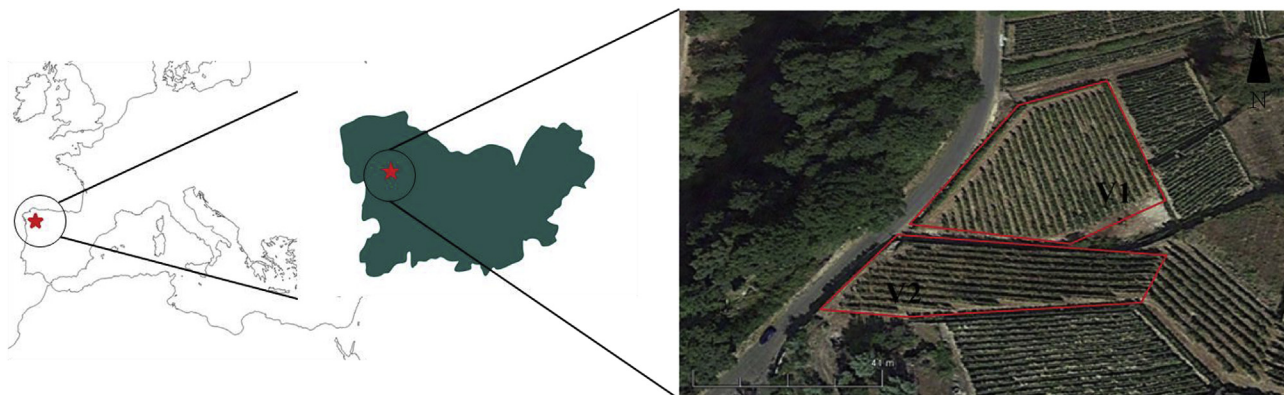


Fig. 1. Location map of the study area and the contours (in red) of the two vineyard plots namely V1 and V2 superimposed over the aerial photograph. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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