Pesticide emission modelling and freshwater ecotoxicity assessment for Grapevine LCA: adaptation of PestLCI 2.0 to viticulture

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Abstract

Purpose Consumption of high quantities of pesticides in viticulture emphasizes the importance of including pesticide emissions and impacts hereof in viticulture LCAs. This paper addresses the lack of inventory models and characterization factors suited for the quantification of emissions and ecotoxicological impacts of pesticides applied to viticulture. The paper presents (i) a tailored version of PestLCI 2.0, (ii) corresponding characterization factors for freshwater ecotoxicity characterization and (iii) result comparison with other inventory approaches. The purpose of this paper is hence to present a viticulture customized version of PestLCI 2.0 and illustrate the application of this customized version on a viticulture case study.

Methods The customization of the PestLCI 2.0 model for viticulture includes (i) addition of 29 pesticide active ingredients commonly used in vineyards, (ii) addition of 9 viticulture type specific spraying equipment and accounting the number of rows treated in one pass, and (iii) accounting for mixed canopy (vine/cover crop) pesticide interception. Applying USEtox™, the PestLCI 2.0 customization is further supported by the calculation of freshwater ecotoxicity characterization factors for active ingredients relevant for viticulture. Case studies on three different vineyard technical management routes illustrate the application of the inventory model. The inventory and freshwater ecotoxicity results are compared to two existing simplified emission modelling approaches.

Results and discussion The assessment results show considerably different emission fractions, quantities emitted and freshwater ecotoxicity impacts between the different active ingredient applications. Three out of 21 active ingredients dominate the overall freshwater ecotoxicity: Aclonifen, Fluopicolide and Cymoxanil. The comparison with two simplified emission modelling approaches, considering field soil and air as part of the ecosphere, shows that PestLCI 2.0 yields considerable lower emissions and, consequently, lower freshwater ecotoxicity. The sensitivity analyses reveal the importance of soil and climate characteristics, canopies (vine and cover crop) development and sprayer type on the emission results. These parameters should therefore be obtained with site-specific data, while literature or generic data that are acceptable inputs for parameters whose uncertainties have less influence on the result.

Conclusions Important specificities of viticulture have been added to the state-of-the-art inventory model PestLCI 2.0. They cover vertically trained vineyards, the most common vineyard training form; they are relevant for other perennial or bush crops provided equipment, shape of the canopy and pesticide active ingredients stay in the range of available options. A similar and compatible model is needed for inorganic pesticide active ingredients emission quantification, especially for organic viticulture impacts accounting.

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1 Introduction

Wine production benefits from a "green industry" image (Brugière 2009; Berghoef and Dodds 2013; Christ and Burritt 2013). Due to the high pest sensitivity of vine, wine industry however applies 13 % in mass of all synthetic pesticides used in Europe, while it occupies only approximately 3 % of the European cropland (Muthmann and Nadin 2007), which is in accordance with observations made in California (Christ and Burritt 2013), where the share of viticulture in terms of pesticide consumption also is larger than its share in agricultural land use. Numerous environmental concerns are related to pesticide use, like surface and groundwater contamination, contaminated runoffs from the fields, bee poisoning (Christ and Burritt 2013) and/or emission of toxic active substances to the air compartment (Ducroz 2006; ATMO Drôme-Ardèche et al. 2010). For these reasons and due to the considerable contribution from pesticide active ingredients (PAIs) to impacts in agricultural products LCAs (Bessou et al. 2012; Godard et al. 2012; Vázquez-Rowe et al. 2012), emissions of PAIs are a key topic to be addressed when performing wine and/or grape production LCAs.

Due to the lack of viticulture-specific inventory models capable of quantifying pesticide emissions and limited availability of characterization factors (CFs) for relevant PAIs, most of the published wine LCA studies neglect toxicological impacts from PAI emissions (Ardente et al. 2006; Gazulla et al. 2010; Bosco et al. 2011; Pattara et al. 2012; Point et al. 2012; Benedetto 2013). Other authors considered substance generic pesticide emission fractions as Neto et al. (2012) such as 25 % to the air and 75 % to the soil or as Petti et al.(2006) who in an LCA of organic viticulture assumes that 50 % of a copper pesticide is absorbed by the plant and 50 % reaches the soil before continuing on to the groundwater compartment (i.e. hence disregarding issues such as drainage system interception of percolate, etc.). Regarding other crops, Nemecek and Schnetzer (2011) assume for all agricultural crop pesticide inventories that 100 % of the applied pesticides are emitted to the soil.

Vázquez-Rowe et al. (2012) and Villanueva-Rey et al. (2014) were the only authors using a substance-specific model to estimate pesticide emissions in wine or wine grape LCAs. Both assessments applied PestLCI 1.0 (Birkved and Hauschild 2006). PestLCI is a dedicated inventory model intended to calculate organic pesticide emissions from arable land (technosphere) to the environment (ecosphere) to be used in (life cycle) impact assessment modelling.

PAI emissions vary and are results of interactions between the properties of the PAIs, the local environment (including meteorology) and agricultural practices (Aubertot et al. 2005). This substance and context dependency is taken into account by PestLCI, which is currently the most advanced LCI model for PAI emissions from agricultural fields (van Zelm et al.

2014). The most recent version of the model, PestLCI 2.0, described in Dijkman et al. (2012) and further modified as described in Dijkman (2014), covers approximately 90 active ingredients of various types of pesticides, 25 European climate profiles and 7 European soil profiles.

Despite the rather extensive coverage in terms of pesticides, climates and soils, PestLCI 2.0 does not take into account certain specificities of viticulture like double cropping system, vertical spraying, and specific PAIs, which differentiate viticulture from other crops and influence the pesticide emission patterns from viticulture compared to other crops. The aim of this paper is to present a tailored version of PestLCI 2.0 customized to appropriately account for the viticulture specificities influencing pesticide emission and to compare the results of this approach to that of other simplified LCI approaches. The approaches compared all have advantages and drawbacks. Table 1 presents the advantages and drawbacks that we have identified for the three inventory approaches compared here.

This paper addresses successively (i) the inclusion of specificities of viticulture in the customized PestLCI 2.0 version, (ii) the development of CFs for freshwater ecotoxicity (FwEtox) using the USEtox™ characterization model for viticulture specific PAIs not covered by the current USEtox CF database, and (iii) the application of the customized inventory model, on a case study of three different conventional[1] vineyard Technical Management Routes (TMRs[2]). The application is further supported and illustrated by characterization of the freshwater ecotoxicological impact potentials through combination of emission quantities and FwEtox characterization, (iv) a sensitivity analysis of PestLCI 2.0 for the identification of the most influential inputs of the model.

2 Methods

2.1 Customization of PestLCI 2.0

In order to improve the viticulture specificity of PestLCI 2.0, the following updates were applied to the model:

- Twenty-nine pesticide active ingredients frequently used in European viticulture
- Thirty-four vine and cover crop development stage combinations
- Nine viticulture-specific pesticide application techniques and corresponding wind drift curves typically employed in French viticulture

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[1] "Conventional" will be used in this paper to designate non-organic plant protection practices.
[2] Technical management routes (TMRs): logical successions of technical options designed by the farmers (Renaud-Gentié et al. 2014)

Table 1 Overview of the advantages and drawbacks that we identified for the inventory approaches compared in this paper

		Pesticide inventory approach		
		PestLCI	Ecoinvent	Neto et al. 2012
Advantage		- Spatial specific	- Easy to apply/applicable by all assessors	- Easy to apply/ applicable by all assessors
		- Temporal specific		
		- Pesticide specific		
		- Application technique specific		
		- Crop specific		
Drawback		- Data (and time) demanding	- Over simplistic/may introduce large uncertainties	- Overly simplistic/may introduce large uncertainties
		- Highly specialized in LCA terms, not necessarily applicable by all life cycle assessors		

- Five Loire Valley soil profiles
- Twenty-two French temperate maritime climate profiles
- The modelling and interpretation of pesticide runoff from the field surface was changed, depending on whether surface water is present near the field.

A summary of these updates is presented in Table S1 in the Electronic Supplementary Material. The customization undertaken is designed for modelling of vertical shoot positioning trained vineyards, which by far is the most frequent training system[3] for vineyards in France and other wine-producing countries. In the remainder of this section, the aforementioned updates are described in more detail. Most of the updates include an expansion of the PestLCI 2.0 databases. The new data included in the model can be found in the Online Resource.

2.1.1 Active substances for pest, diseases and weed management in viticulture

An average number of 16 pesticide active ingredients (PAIs) were applied to French vineyards in 2010 (with high interregional variability). Downy and powdery mildew fungi were the target pests in 95 % of the 12 applications (Ambiaud 2012b).

A variety of PAIs are registered for viticulture farming in Europe, from generic farming PAIs to more crop-specific PAIs shared with pest management in vegetables or in orchards. The latter pesticide types were not available in the original PestLCI 2.0 version. Hence, on the basis of the list of the viticulture-specific PAIs applied over four vintages (2010 to 2013) (see Electronic Supplementary Material, Table S2), compilation of data on the properties of the relevant organic PAIs used in viticulture was conducted applying dedicated chemical/fate property databases (refer to the Electronic Supplementary Material, Sect. S-C and Table S3 for a more thorough introduction to the missing viticulture relevant PAIs in PestLCI 2.0).

Inorganic fungicides based on copper and sulphur are widely used in viticulture, especially organic viticulture (see more details on vine pests and diseases management, copper and sulphur in the Online Resource, Sects. S-A and S-B). Sulphur represented, in 2003, 69 % in mass of the PAI applied in the European Union on vineyards, and cupric compounds, 2.7 % (Muthmann and Nadin 2007). Conventional viticulture also uses other inorganic PAIs such as ammonium thiocyanate (herbicide) or partially inorganic PAIs like fosetyl-Al (fungicide). However, inorganic or partially inorganic substances behave and react differently compared to entirely organic[4] pesticide due to speciation. Their emission loads can hence not be modelled, as organic pesticides, applying PestLCI 2.0. For this reason, these types of PAIs were not included in this study.

In addition, more "exotic" PAIs were likewise not considered in the present study. This third PAI group includes the following:

– PAIs not officially approved/registered as pesticides such as algae extracts (only registered as fertilizers)
– Pesticide formulation additives (e.g. light paraffinic oil, canola oil, glycerol and lignite), due to lack of information about their properties and occurrences in the assessed pesticides, despite the fact that these substances can contribute considerably to toxicity of the pesticide formulation (Brausch and Smith 2007) and modify PAIs drift potential (Celen 2010)

2.1.2 Spraying equipment for application of pesticides

PestLCI 2.0 takes into account the type of sprayer applied for the application of the pesticide in order to quantify the drift

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[3] Training system: type of trellis and shoot positioning resulting to a given shape of the vine canopy and position of grapes.

[4] "Organic" is alternately used in the paper to qualify a type of crop management which uses no synthetic pesticides and a chemical type of PAIs: organic chemical compounds containing covalent bound carbon, oppositely to inorganic chemical compounds (inorganics) which do not contain carbon bound this way. Here, "organic" relates to the chemical compound nature.

through drift curves. The types of spraying equipment applied in viticulture are numerous, which makes the task of modelling the individual equipment characteristics a challenge. The sprayers designed for canopy and grapes spraying may use different modes of droplets production: non air-assisted spray, air blast and pneumatic. Different shapes of the ventilators and of the sprayers themselves lead to different patterns in terms of spraying quality and drift generation.

None of the above presented culture-specific application techniques were available in PestLCI 2.0. In the present customization of PestLCI 2.0, nine new viticulture-specific sprayers were included. The nine sprayer types are described in Table S7 in the Electronic Supplementary Material. Of these, a tunnel sprayer is based on data by Ganzelmeier (2000) and eight items from Codis et al. (2011), who published the only drift measurements obtained in France for vineyards according to the ISO protocol (ISO 2005). We assumed that the bias caused by the vine rows width difference between the test setup of Codis et al. (2011) and our modelling approach (1.40 m compared to ours are 1.90 to 2.50 m) would lead to smaller uncertainties than relying data for non-viticulture-specific spraying equipment. From the results of these nine drift measurements, drift curves were derived. These are given in Table S8 in the Electronic Supplementary Material. A user guide for the choice of sprayer type will soon be available for the users of PestLCI 2.0.

According to the design of the sprayer, wine growers can choose to spray one to four rows of vines simultaneously. The number of rows treated plays a significant role in wind drift calculation in PestLCI 2.0. This issue has been taken into account by entering the actual width treated at the same time along with the parameter "nozzle distance" in the model.

Herbicides are most often applied very close to the soil with specific sheltered booms to avoid herbicide drift and hence deposition on vine leaves. We chose to model this application technique as the existing "soil incorporation" in PestLCI 2.0 since sheltered boom sprayers induce very low drift.

Finally, modelling of custom spray techniques covering various adaptations of existing spraying equipment is considered beyond the scope of this paper.

2.1.3 Accounting for primary distribution in double cropping systems

Cover cropping on vineyard soil is a developing management scheme with nearly half of the French vineyards temporarily or permanently applying double cropping (Ambiaud 2012a). A second canopy under the vineyard (e.g. spontaneous species, oats, clover or fescue) can cover various proportions of the row width and present various densities. The secondary crop contributes to pesticide interception (primary distribution) and fate (secondary distribution), which increases the

pesticide's potential for volatilization while limiting runoff from topsoil.

The primary distribution process is defined in PestLCI by three fractions: wind drift (f_d), pesticide deposition on soil (f_s) and pesticide deposition on leaves (f_l) (Birkved and Hauschild 2006). The two latter are based on Linders et al. (2000) interception fractions for single crops at different development stages. In terms of interception by the vine canopy, PestLCI 2.0 includes interception values for vine at four different development stages I, II, III and IV based on Linders et al. (2000). We added an additional stage 0 in PestLCI 2.0 in order to take into account situations of leafless vines (see Online Resource S-D for details). We further adjusted vine interception fractions by considering results of on-field measurements of spraying mixture deposition and losses on vineyards by Sinfort (pers comm 2014) and Sinfort et al. (2009) and on artificial vineyard (test bench reproducing the shape of a vineyard where the leaves are replaced by papers for droplets quantification) by Codis et al. (2014). Distribution fractions of spray mixtures between vine canopy, soil and air at 2.5 m above the soil were obtained by these authors in vineyard conditions similar to the ones we study (rows width, types of sprayers). The fraction sent to air during an application measured by these authors was introduced in PestLCI 2.0 as being (i) partly conveyed by wind drift out of the parcel (i.e. advective transport) and (ii) partly falling back on vegetation and bare soil of the parcel (i.e. sedimentation). This choice was made because no quantification of direct volatilization during spraying is possible (Jensen and Olesen 2014) due to the complexity of volatilization driver combinations (properties of the spray liquid, drops size and drops surrounding conditions) (Gil et al. 2008) and the lack of available data for some of the equipment-specific parameters. The details of these drift calculation including equations are available in the Online Resource Sect. S-D.

The interception by the cover crop, as modelled in the version of PestLCI 2.0 presented in this work, varies according to the width of the cover crop strips estimated as a percentage of the width of the vine inter-row and according to cover crop canopy density (see Fig. 1a–c).

A consequence of this change in emission modelling compared to a situation in which cover crop is not present is that, in the initial distribution, less pesticide will reach the soil and more will be present on vine and grass leaves, meaning the fraction intercepted by the crop canopies increases compared to monocultures. As a consequence, less runoff of dissolved pesticide and volatilization from top soil should be expected. On the other hand, more pesticide can be expected to volatilize from the leaves of the cover crops. In general, volatilization rates are higher from leaves than soil, so for most pesticides, an increase in emissions to air can be expected.

Combined interception factors for mixed canopies (vine + cover crop) were included in the model for the most typical

Fig. 1 **a**–**c** Vine I grass 0 %, vine I grass 100 % average density, and vine IV grass 50 % high density (pict. 1 and 2, E Bezuidenhoud, pict.3: P. Rodriguez-Cruzado)

situations as the following product: [vine development stages×cover-crop strip width×grass canopy density] (see Table 2).

2.1.4 Climate and soils datasets

Site-specific climatic profiles appropriately representative for the case study areas were included in PestLCI 2.0. To permit sensitivity tests on climate data, two sets of 30 years average 1971–2000 and 1981–2010 for the Beaucouzé Station were added to PestLCI, as well as for five stations of the Middle Loire Valley, located close to the studied vineyards. For these five stations data for 3 years of production, i.e. October year n to September year $n+1$, for 2009–2010 to 2011–2012, as well as sets of average months for the 3 years are available see Table S5 in the Electronic Supplementary Material. Climatic data were provided by "Météo France". Five soils corresponding to the modelled parcels were characterized through measured data and observations, in accordance with the PestLCI 2.0 data requirements, and entered in PestLCI 2.0. (see Table S6 in the Electronic Supplementary Material).

2.1.5 Modelling of pesticide runoff from the field surface

The modelling of buffer zones around the field was altered. In previous versions of PestLCI, the width of the buffer zone was fixed, independent of both the presence of surface water, which these zones are intended to protect, and the distance to this surface water. In the updated model, the user can

indicate whether a freshwater body is located near the field. If this is the case, the user has to specify the distance to the water body. In case this distance is less than the required buffer zone around the field, a part of the field will be considered a part of the buffer zone between the area undergoing pesticide application and the freshwater body. If there is no water body nearby, any surface runoff from the field will be considered as an emission to the soil outside the field; therefore, a compartment was added: nearby agricultural soil. Soil was chosen as an emission compartment, because this compartment better represents the fate of the pesticide than other environmental compartments. When surface water is not nearby, the runoff water will end up on or in the soil, and the pesticide will partition between the soil solid matter and the air and water in the soil pores. Emissions to this compartment were characterized as emissions to continental agricultural soil in USEtox™.

2.1.6 Calculation of USEtox™ CFs

CFs are needed in LCA to quantify the potential environmental impacts resulting from emissions occurring over the life cycles of products and systems. CFs are generally substance and compartment specific and sometimes spatially explicit since the impact pathways of an emission depends on the substance, the emission compartment and to some extent the geographic location of the emission. In this study, we used CFs obtained from the USEtox™ characterization model since the model was developed as a scientific consensus

Table 2 Examples of combined interception factors for vine/cover crop mixed canopies (complete table available in the Electronic Supplementary Material, Table S4)

Stage	Density of cover crop canopy	% of soil surface covered by cover crop	f_{vine}	$f_{covercrop}$	% spray intercepted by vegetal soil cover (calculation)	f_{Ic}
0	None	0	0.1	0.3	0 %	0.10
II	Weak (30 %)	100 %	0.5	0.3	6 %	0.56
II	High (70 %)	80 %	0.5	0.7	11 %	0.61
III	Average (50 %)	100 %	0.65	0.5	5 %	0.70

model, supposedly representing the best application practice for characterization of toxic impacts of chemicals in LCA (Hauschild et al. 2008) and since its database (v. 1.01) covers ~2500 chemicals with calculated CFs for FwEtox (Rosenbaum et al. 2008). USEtox™ is not spatially resolved but operates with a nested structure that distinguishes between an urban (air compartment only), continental and global scale.[5] Following common practice, we in this study applied CFs from the USEtox™ database (v. 1.01) for emissions to the continental air, agricultural soil and freshwater compartments. Of the 48 PAIs covered by this study, the default USEtox™ database currently does not cover 21 (see Table S2 in the Electronic Supplementary Material). To fill these gaps, we applied the USEtox™ model to calculate CFs for emissions to the continental air and freshwater compartments for the 18 organic PAIs of the 21 PAIs missing in the default database (the USEtox™ model is not designed to characterize inorganic emissions; hence, three inorganic PAIs were left out). Leaving out these three pesticides will have some effect on the results; however, lacking emission and characterization data on the three substances left out obstruct assessment of the errors introduced hereby.

Due to the considerable contribution to the total impact score from Folpet and the calculation of a much lower CF by AiiDA (Hugonnot et al. 2013), we recalculated the CF for Folpet based on best available data. We found that input parameters related to physical-chemical properties of the PPDB (University of Hertfordshire 2013) database were generally of a higher quality (more experimental values) than the data from the EPISuite (US Environmental Protection Agency 2012) used in the calculation of the Folpet CF from the default USEtox™ database. We therefore recalculated CFs based on PPDB input data (where these were available) for physical-chemical properties but did not change "avlogEC50" (the input parameter for ecotoxicity), since this parameter was based on test data from 26 species, representing 4 trophic levels and therefore deemed to be of a high quality. The input data used for recalculating the CFs of Folpet and the resulting set of CFs are presented in Table S9 and Table S10 in the Electronic Supplementary Material.

Since USEtox™ is spatially generic, these new CFs may be applied to case studies anywhere in the world. The calculations followed the procedure of the USEtox™ manual. Experimental data inputs were prioritized over modelled data inputs (see Tables S9 and S10 for data sources and data used, Electronic Supplementary Material). Regarding uncertainties of the calculated CFs, we followed the classification of the USEtox™, which flags CFs as "interim" if a number of criteria for (relatively) low uncertainty are not fulfilled.

2.2 Case study

Three contrasted conventional TMRs of *Chenin Blanc* cultivar in the Middle Loire Valley (France), studied during 2010–2011 production year, were chosen to illustrate the applicability of the PestLCI 2.0 customization for viticulture and new USEtox™ CFs. The cases presented here are part of a project aiming to establish a method for joint evaluation of environmental (through LCA) and qualitative performances of viticultural TMR (Renaud et al. 2012).

2.2.1 Functional unit

The emissions and impacts calculated in our paper are presented per ha because vine, as a perennial crop, occupies land for several decades (sometimes centuries) and vineyards in addition have an important function of maintaining space and landscape values (Joliet 2003; Renaud et al. 2012). Moreover, this functional unit accounts for the goal of minimizing the impacts while cultivating a given area (Mouron et al. 2006), and it is hence considered more adequate for communication towards winegrowers who typically reason in terms of farming management practice per ha. The emissions and impacts can be calculated per kilogram of grape, by dividing the results by the yield of each parcel.

2.2.2 Geographical situation, cultivar and practices

The Middle Loire Valley's cool and sub-humid climate (Tonietto and Carbonneau 2004) offers favourable conditions for growing different sorts of vine (*Vitis vinifera*) cultivars and producing a wide range of wine types in more than 50 different wine production areas labelled "Protected Denominations of Origin" (PDO[6]). Chenin Blanc is the typical and the main white cultivar of this area, used to produce dessert-style sweet, dry and sparkling white wines. The three vineyard TMRs chosen for the present study are designed for PDO Chenin Blanc dry wine production in the PDO zones Anjou Blanc and Saumur Blanc. The soils and subsoils of the Anjou PDO zone are mainly schist and metamorphic sandstone of the Armorican Massif, while the Saumur PDO zone is located on the sedimentary marl, chalk and calcareous sands of the Parisian Basin (Goulet and Morlat 2011). Despite the PDO set of rules fixing some practices like training system or rows width (similar for the PDOs represented in the present survey), an important diversity remains for the other practices. The three TMRs studied are all represented by real vineyard situations. The choice of these three real situations was based on

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[5] USEtox™ contains no ground water compartment. Ecotoxicological impacts in freshwater from chemical emissions to groundwater are considered negligible and thus not further considered in this study.

[6] PDOs promote and protect names of quality agricultural products and foodstuffs which are produced, processed and prepared in a given geographical area using recognized know-how (European-Commission 2014).

the results of a regional survey analysed according to Typ-iti method (Renaud-Gentié et al. 2014), in order to represent the diversity of vineyard management of Chenin Blanc grown for PDO dry white wines production in Middle Loire Valley. Five types of vineyard TMRs emerged from this survey analysis: (1) "systematic synthetic chemical use and limited hand-work", (2) "moderate chemical use", (3) "minimum synthetic treatments and interventions (i.e. mechanical or manual oper-ations)", (4) "moderate organic" (i.e. with limited interven-tions and treatments), and (5) "intensive organic" (i.e. with many interventions and treatments). All five TMRs are further described in Renaud-Gentié et al.(2014). The cases studied in the paper at hand concern practices of the winegrowers ob-served on three plots representative of the three first TMR type, and the two last TMR types are organically managed and thus involve nearly exclusively inorganic PAIs which are not modelled in PestLCI 2.0.

2.2.3 Climate of the studied year

The results presented here relate to production year 2010–2011 (October 1, 2010–September 30, 2011). Based on the Angers-Beaucouzé weather station (main station of the area) data, the production year 2010–2011, in comparison to the average of 30 years 1981–2010 (Fig. 2), 2011 can be de-scribed as follows: (i) a little warmer (+0.2° on the annual average) with a warmer spring but a cooler July, (ii) much drier especially during the vine growing season (−60-mm rain and +40-mm potential evapotranspiration in the April–September period on an average total of 306-mm rain for this period and 657.4-mm potential evapotranspiration).

The particularly low precipitations in spring may generate lower emissions to groundwater, and the higher temperatures can cause higher emissions to air than an average year. We performed a sensitivity analysis on these climatic inputs.
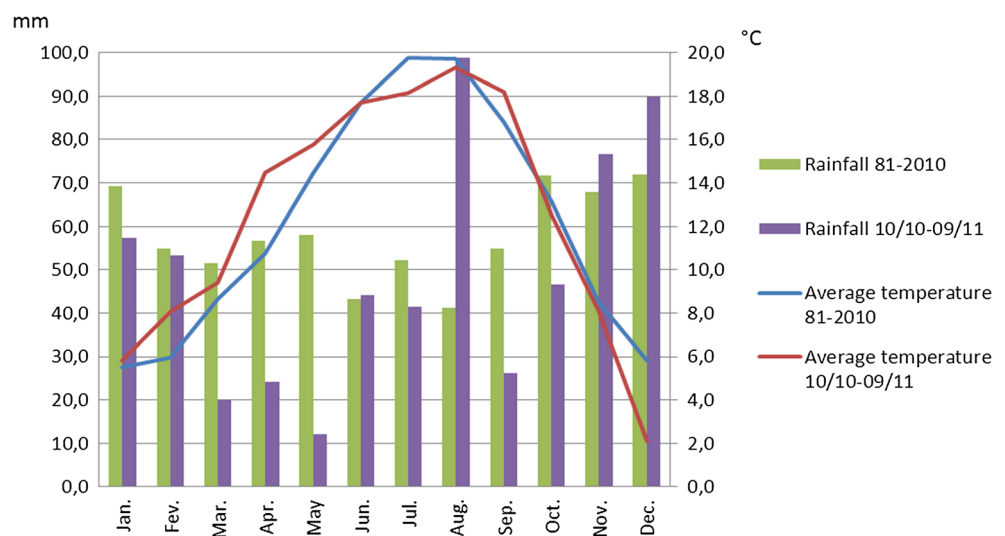
2.2.4 Soils, environment and yields

Each plot presents a different type of soil but quite similar slopes (3 to 6 %). The soil layers were described by field observation with soil auger and soil analysis, and consolidated with comparison to existing detailed soil cartography of vine-yard soils of the Middle Loire Valley. The soil characteristics were implemented in the PestLCI 2.0 soil database. Table 3 summarizes the soil characteristics of the three studied TMRs' plots. Soil characteristics and tillage should play a role on emissions to groundwater by changes in soil porosity. Slope and drainage should influence emissions to surface water, as should cover crop extent, and the latter should additionally influence emissions to air by changes in canopy area. The sensitivity analyses will explore the influence of soil, slope, tillage and cover-crop extent parameters on the results.

No surface water body lies at less than 100 m from the parcels. The plots are not drained. They are all cover-cropped, but the covers present different densities and extents. Irrigation is not allowed in PDO vineyards under Middle Loire Valley climate; hence, the studied plots are not irrigated (irrigation water would have to be added to rainfall and thus increase surface water emission rate). The yields for 2011 were the following: TMR1 8000 kg grapes/ha, TMR2 5250 kg grapes/ha and TMR3 7500 kg grapes/ha.

2.2.5 Vineyard protection programs

For each TMR, different spraying equipment and PAIs were used by the growers (see Table S12 in the Electronic Supplementary Material). Defining which of the nine sprayers added to PestLCI 2.0 is most similar to the sprayers used by the growers was done through discussion with S. Codis (pers. comm. 2014). Since the chosen sprayer type determines pesticide drift, which may influence the modelled emissions to

Fig. 2 Main characteristics of the climate of production year 2010–2011

Table 3 Soil and cover crop characteristics of the three TMR studied

Case	Soil	Slope%	Cover crop extent	Tillage
TMR1	- UTB131	5	70 % high density	No
TMR2	- UTB25	6	30 % average density	No
TMR3	- UTB35	3	50 % average density	No

air, the choice of sprayer type is included in the scenario uncertainty analysis.

2.3 Sensitivity analyses

Two types of sensitivity analyses were carried out in order to identify the parameters towards which the outcomes of our customized version of PestLCI 2.0 are most sensitive and hence which parameters should be focused on to reduce uncertainty caused by inventory work and landscape parameters documentation in future studies. Input parameter sensitivity (on quantitative parameters) and scenario sensitivity analysis (on qualitative parameters) were conducted.

The input parameter sensitivity analysis was carried out for the application of Folpet in TMR1. Folpet was chosen for this analysis, because it is the organic PAI the most frequently used in viticulture in France (Ambiaud 2012b). As can be seen from Table S12 in the Electronic Supplementary Material, in TMR1, Folpet is applied in May using a recycling tunnel. The vineyard measures 100×100 m, the soil of UTB 131 has a slope of 5 % and it not drained. There is no surface water near the vineyard; therefore, runoff of dissolved pesticide is classified as an emission to agricultural soil. The climate used to model this scenario was Blaison-Gohier's. Starting from this basis scenario, 37 parameters were, one at a time, increased with 10 %. These parameters include direct inputs that can be modified by PestLCI 2.0 users, as well as parameters included in the model's climate and soil profiles and properties of the active ingredient. Each parameter was changed with the same percentage in order to allow for a comparison of the sensitivities of the different parameters. For each change in input parameter, the emissions to air, agricultural soil and groundwater were calculated. Finally, the percentages of change in the emissions were calculated. Since the aim of this assessment is to focus on the inventory data collection, rather than determining the sensitivities of the final results, this sensitivity assessment was carried out for one active ingredient.

The scenario sensitivity analysis was conducted on the inputs that involve discrete data, i.e. type of sprayer, of soil or climatic datasets. The effects of input change on the model outputs were assessed in terms of percentage of variation of the output in comparison to a reference case. The tested input types were assessed on basis of the same PAI application event, by varying one parameter at a time. A reference case was chosen for each input type (Table 4). For example, the tunnel sprayer was taken as the reference sprayer, and the emissions found for the other sprayers were expressed as a negative or positive change of the emissions, expressed in a percentage, compared to the emissions calculated with the tunnel sprayer.

3 Results

3.1 Case study: emissions of organic PAIs and FwEtox

3.1.1 With Pest-LCI 2.0

Emissions were calculated by PestLCI 2.0 for every organic substance application done in 2011 for the three TMRs. Inorganic PAIs were excluded from the calculation, since they fall outside the scope of PestLCI 2.0.

The emission fractions vary to a large extent. These variations are determined by the PAIs' properties as well as parcel and application conditions (Fig. 3).

They do not exceed 0.35 and are lower than 0.15 for most of the PAI applications. They are highly dominated by air emissions, followed by ground water emissions. Emissions to nearby agricultural soil are negligible (from 2×10^{-20} to 2×10^{-4}) and thus not visible on the charts. The absence or quasi-absence of freshwater emissions can be explained by the absence of water body around the parcels.

The three fungicides Tetraconazole, Cymoxanil and Mefenoxam were found to have the highest emissions, followed by two herbicides (Aclonifen and Amitrole).

For a same PAI, e.g. Amitrole, sprayed in all three TMRs, with the same type of boom, and on the same canopy (grass), emissions to air and to groundwater vary because of different soil and climatic conditions. These drivers are explored in the sensitivity analyses section.

High emission fractions do not necessarily lead to high emissions: For most of the PAIs, high emissions are compensated by very low application doses (Cymoxanil, Tetraconazole), leading to moderate emissions quantities (Fig. 4).

The quantity of PAIs emitted per application is not higher than 0.14 kg/ha in all scenarios. As it was the case for the emission fractions, the emission quantities are dominated by air emissions. Due to the combination of a large quantity applied (around 1 kg/ha) and high emission fractions, Amitrole dominates the emissions to air in the three TMRs. After, Amitrole, Folpet and Aclonifen show the highest emissions. In contrast, for Mancozeb, though applied at high rates, moderate emissions are observed due to low emission fractions.

FwEtox calculated applying USEtox™ CFs (Fig. 4) reveals high differences for the different applications, due to high disparities in ecotoxicological profiles of the PAIs. The

Table 4 Tested input types for scenario uncertainty analysis, reference characteristics and number of alternatives tested

Tested input "type"	Reference	PAIs	Month	Alternatives tested
Weeding booms	PestLCI 1 soil incorporation	Aclonifen	March	IMAG conv boom bare soil, IMAG conv boom cereals
Sprayers	Tunnel sprayer	Folpet	May	sprayer IDK, sprayer spider vault, sprayer CG pneumatic, sprayer Abmost pneumatic, sprayer GRV fantip, sprayer GRV AVI air assisted, sprayer GRV AVI non air assisted, sprayer pendillard TVI, sprayer crossflow fruit
Interception by mixed canopies	Vine 0 0 %grass	Folpet	March	Vines 0—w30% grass, Vines 0—h30% grass, Vines I—a0% grass, Vines I—w50% grass, Vines 0—h80% grass, Vines II—a0% grass, Vines II—w100% grass
Soils	UTB 131	Folpet	March	UTBs 11, 25, 35, 156
Tillage	No tillage	Folpet	March	Tillage
Months	March	Folpet	March	April, May, June, July, August
Climatic dataset	Oct. 2010: Sept. 2011	Full program (11 PAIs)	March: July	10/2009–9/2010; 10/2011–9/2012; average of the 3 years10/2009–9/2012; 30 year average 1981–2010 Beaucouzé

FwEtox of TMR1 is dominated by Aclonifen (500 PAF m^3 day), Fluopicolide (80 PAF m^3 day) and Cymoxanil (40 PAF m^3 day). The other TMRs show much lower FwEtox than TMR1.

Multiple factors differentiate the case vineyards TMR1, TMR2 and TMR3. The main factors are considered to be soil characteristics, sprayer equipment used and type of pesticides applied. TMR1 shows higher emission fractions than TMR3; however, the total mass of emitted pesticide is lower because of the low doses applied for some substances. TMR2 and TMR3 show a much lower total FwEtox (33 and 37 PAF m^3 day) than TMR1 (634 PAF m^3 day), mainly due to the high ecotoxicity of Aclonifen used in TMR1, even if this PAI is applied via sheltered boom, limiting wind drift. The comparison between the three TMRs discussed here considers only organic PAIs, even though inorganic substances are also involved in these three vine protection strategies but could not be assessed.

3.1.2 Comparison of PestLCI 2.0 results with two simplified emission modelling approaches

The Ecoinvent approach applied for pesticides assumes that 100 % of the applied pesticide is emitted to the soil (Nemecek and Schnetzer 2011); thus, the agricultural soil is considered part of the ecosphere. Neto et al. (2012) in their LCA of Portuguese wine Vinho Verde propose a substance generic partition as with 75 % of pesticides emitted to soil and 25 % to the air. The results between the three approaches were compared on TMR1 to TMR3 organic pesticides application program (Fig. 5).

As the results are not normally distributed, means and standard deviation cannot be used; results are thus compared through their medians and their distribution.

In the present study, the median of total emission fraction modelled with PestLCI 2.0 is 26 times lower than the total emission fractions estimated by the Ecoinvent and Neto et al.
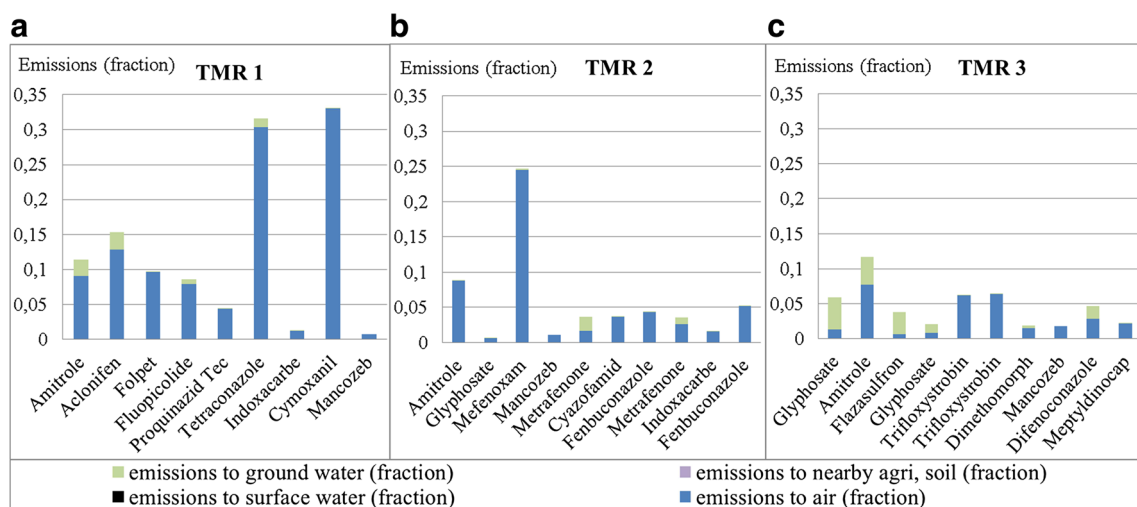
Fig. 3 a–c Fraction of applied PAIs emitted in the four compartments presented in the chronologic order of application during 2011 cultivation year
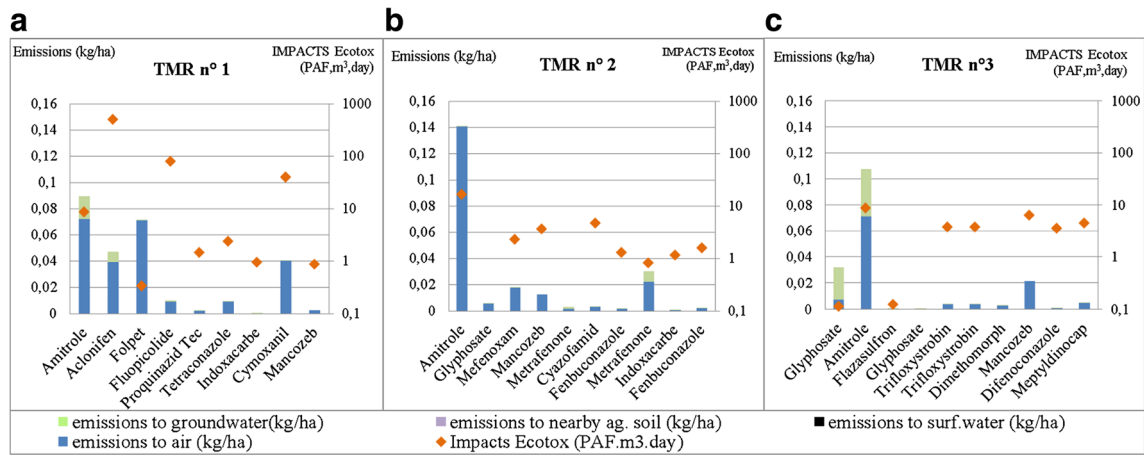
Fig. 4 **a**–**c** Quantities of PAIs emitted and per hectare of vineyard in the four compartments and FwEtox calculated by USETox™ (note the log scale for FwEtox impacts) in the chronologic order of application during the 2011 cultivation year

(2012) approaches (Neto et al. (2012) total emissions= 25%air+75%soil=100 %=Ecoinvent soil emissions). The median of PestLCI 2.0 modelled emission fraction to air is seven times lower than the total emission fraction to air estimated by the Neto et al. (2012) approach.

This leads to huge differences in FwEtox estimates (USETox™ CFs applied in all cases) (Fig. 6): 32 times lower with PestLCI model than Ecoinvent and 36 times lower than Neto et al. (2012) approach.

Very high variability in FwEtox results within each of the three approaches must be noticed, which can be explained by large differences in the PAIs' CFs.

The emission quantities of individual PAIs that are estimated by PestLCI 2.0 are always lower than the simplified emission modelling approach estimates (Fig. 7).

The PestLCI approach results in total emissions that are between 3 (Cymoxanil, TMR1) and 143 (Glyphosate, TMR2) times lower than the 100 % emitted to soil approach (Ecoinvent). PestLCI emissions to air are between 0.75 (Cymoxanil, TMR1) and 42 (Flazasulfuron, TMR3) times

lower than Neto et al. (2012) approach. Moreover, the ranking of the PAIs on basis of their FwEtox is not the same between PestLCI 2.0 and the two simplified emission modelling approaches.

3.2 Sensitivity analysis

3.2.1 Sensitivity of the model to quantitative inputs

The results for the sensitivity analysis are summarized in Table 5. This table lists the three input parameters to which the emissions to air, surface water and ground water are most sensitive. The sensitivities of all tested parameters are found in Table S13 in the Electronic Supplementary Material.

The emissions to air are mostly sensitive to parameters that determine pesticide presence on leaves like solar irradiation, which affects the rate of degradation. Since degradation competes with volatilization, a change in the degradation rate affects the rate of volatilization. The average ambient temperature affects both the volatilization and degradation rate. The third

Fig. 5 Comparison of PAI emissions and their distribution calculated on the three plots vineyard protection programs (organic PAIs) by PestLCI 2.0, Ecoinvent and Neto et al. (2012) approaches. Each *boxplot* shows the median of all values (*bold line*) flanked by the first (*bottom*) and the third (*top*) quartiles (limits of the *box*) and first (*bottom*) and ninth (*top*) deciles (*whiskers*), outliers are plotted as individual points; three major contributing PAIs are illustrating the differences (*colour points*)
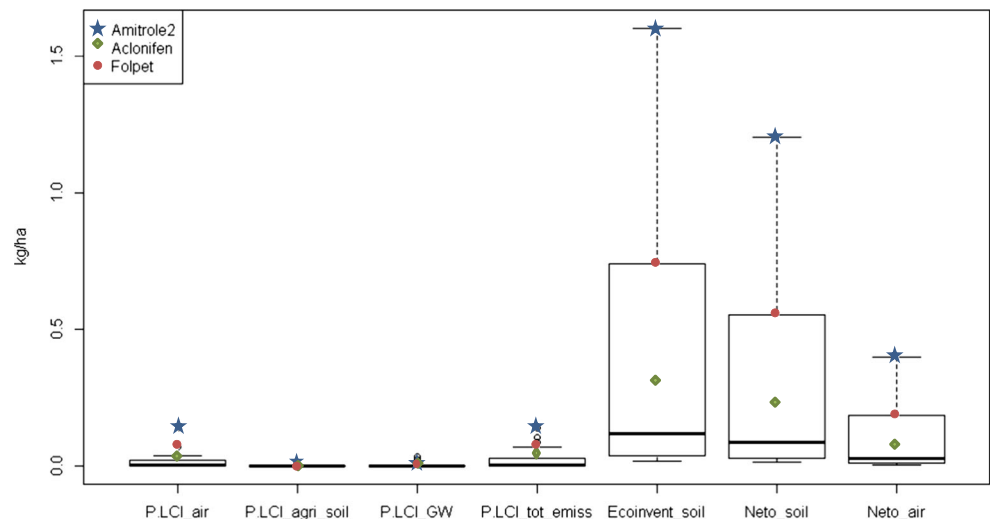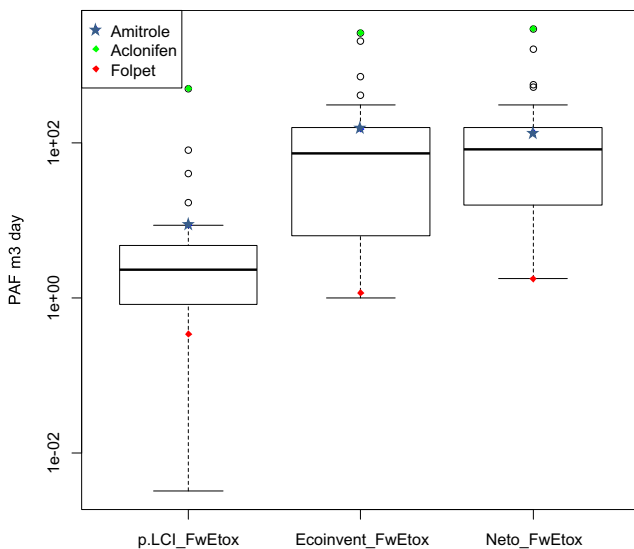
Fig. 6 Comparison of FwEtox calculated on the three TMR's vineyard protection programs emissions (organic PAIs) with USETox™ CFs (logarithmic scale). Each *boxplot* shows the median of all values (*bold line*) flanked by the first (*bottom*) and the third (*top*) quartiles (limits of the *box*) and first (*bottom*) and ninth (*top*) deciles (*whiskers*), outliers are plotted as individual points, and three differently contributing PAIs are illustrating the differences (*colour points*)

most sensitive parameter was found to be the primary interception fraction, determining the pesticide distribution between leaves and soil. The choice of application method can be even more influential than the other parameters tested in Table 5, but as a discrete choice, it was included in the scenario sensitivity analysis (see Sect. 3.2.2). The emissions to nearby agricultural soil (or surface water, had that been present) are sensitive to parameters that determine how much pesticide is present on the soil surface such as the fraction of applied pesticide that is intercepted

by leaves and the soil half-life of the pesticide. Moreover, the slope of the field was shown to be an important parameter: The steeper a slope, the more rain water will start to run off. Finally, emissions to ground water were also found to be mostly sensitive towards the fraction of pesticide that initially reaches the soil, as well as towards soil properties.

3.2.2 Scenario sensitivity analysis

The sensitivities of f_{air}, f_{sw}, f_{gw} and FwEtox to the different inputs cited in Sect. 2.3 were calculated by making each input vary in the range of values available in the model (Table 6).

Sensitivity analysis results of f_{air} and FwEtox show a very strong correlation (see Fig. S1 in the Electronic Supplementary Material) because f_{air} is the major emission route in this case study. For this reason, only f_{air} sensitivity results will be presented in the section below.

The most influential parameters on f_{air} are the interception by the canopy (or canopies) and, to a lesser extent, the climatic annual dataset. Concerning f_{gw}, the main drivers turned out to be the climatic dataset (climatic year or climatic month).

A complementary sensitivity scenario analysis on four climatic dataset including averages on 30 years on a complete treatment program is available in the Electronic Supplementary Material, Sect. S-E.

4 Discussion and outlook

4.1 Case study insights

When using the original USEtox™ CFs for Folpet, the dominancy of Folpet found in the FwEtox results of the
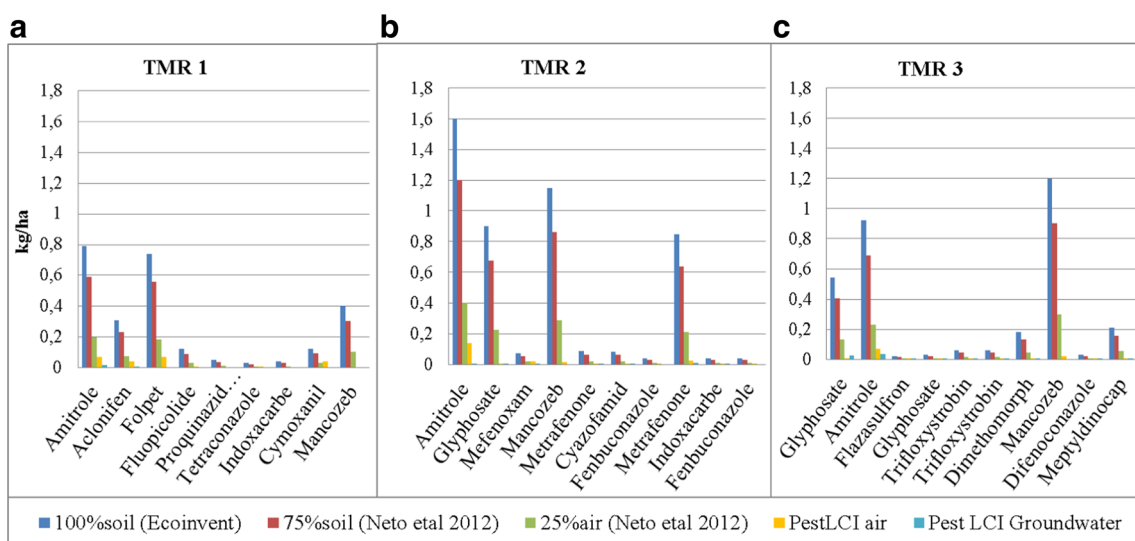

Fig. 7 a–c Comparison of emissions per hectare treated from PestLCI 2.0 and two simplified emission modelling approaches

Table 5 Summary of sensitivity analysis, showing sensitivities as the change in emissions resulting from a 1 % change in the given input parameter

Parameter	Sensitivity (%)
f_{air}	
Solar irradiation	−3.0
$T_{average}$ in the month of application	2.2
Interception fraction	0.99
$f_{sw/ag.soil}$	
Interception fraction	−6.9
Field slope	1.3
Soil half life	1.1
f_{gw}	
Interception fraction	6.9
Soil solid matter fraction	3.2
Soil water fraction	2.1

f_{air} emissions to air, $f_{sw/ag.soil}$ emissions to surface water/near-field agricultural soil, f_{gw} emissions to ground water

present case study is consistent with results obtained by Vázquez-Rowe et al. (2012) and Villanueva-Rey et al. (2014) with PestLCI 1.0, where FwEtox is found to be dominated by Terbuthylazine (which was not applied here, its use being forbidden in France since 2003) and Folpet. A comparison of the present TMRs FwEtox profiles (using the original USEtox™ CFs for Folpet) with the results obtained by Vázquez-Rowe et al. (2012) with PestLCI 1.0 in Galician vineyards shows very good environmental performance of the present TMRs: TMR1's FwEtox is half of the lowest FwEtox mentioned by this author (Copper impacts removed). However, the version of PestLCI used by these authors is an older version and was not customized for viticulture. This may have caused overestimations of the emissions: The recycling tunnel sprayer used to apply Folpet results in emissions to air

that are lower than other application methods available in PestLCI 1.0. Moreover, the emissions to surface water are in general found to be lower in PestLCI 2.0 than in PestLCI 1.0 (see for example Dijkman et al. (2012)). The new CFs that we have calculated for Folpet and used in this paper yield a low FwEtox for this PAI and thus a lower FwEtox for TMR1.

Inorganic or partially inorganic PAIs could not be modelled here because of the lack of model appropriated to their specific physical-chemical behaviour; however, they were also applied to the case vineyards (see Table S12 in the Electronic Supplementary Material): one (TMR3) to five (TMR1) PAIs applications. The copper-based PAIs are particularly expected to further increase the FwEtox of the TMRs if included (Mackie et al. 2012; Vázquez-Rowe et al. 2012). Their widespread use in viticulture reveals the need for models capable of quantifying inorganic PAIs emissions.

4.2 Sensitivity and inventory priorities

The results of the sensitivity analysis shown in Table 5 do not give the same hierarchy between the parameters as those presented by Dijkman et al. (2012). This can be explained by differences in active ingredients, soil, climate and pesticide application methods used as inputs between both studies. In addition, modelling of some of the fate modules in PestLCI has been modified, as described in Dijkman (2014).

The sensitivity analyses show that climate, canopy interception and soil granulometry play major roles in the results of both PAI emissions and FwEtox. Therefore, these parameters should, ideally, not be estimated by default or average values. Moreover, efforts should be put on main contributors to f_{air}, f_{sw} and $f_{ag.soil}$ sensitivity because, in the current state of characterization methods, emissions to ground water are not taken into account for impact calculation.

Table 6 Highest variations of emission fractions per input type

Input type	Reference	PAIs	Highest variation f_{air} in %	Highest variation f_{sw} in %[a]	Highest variation f_{gw} in %	Number of alternatives tested
Weeding booms	PestLCI 1 soil incorporation	Aclonifen	4	−0.53	−0.53	2
Sprayers	Tunnel sprayer	Folpet	51	No emissions	−5	9
Interception by mixed canopies	Vine 0 0 %grass	Folpet	378	−77	−77	7
Soils	UTB 131	Folpet	0.03	−100	−64	4
Tillage	No tillage	Folpet	0	0	−87	2
Months	March	Folpet	43	−63	−73	5
Climatic dataset	Oct. 2010: Sept. 2011	11 PAIs	65	NA	443	3

[a] A freshwater water body was considered at 20-m distance from parcel boundary, except for climatic dataset test

The importance of pesticide interception by plant and cover-crop canopies, especially on f_{air}, implies that width and density of grass cover strip as well as vine development stages must be well documented in viticulture.

The importance of the climatic dataset on emissions to f_{air} and f_{gw} points out the necessity to use the actual climatic dataset of a given year when one wants to assess a real TMR in that given year: the use of another climatic year or long-term average climatic data can introduce important uncertainty in the results.

The choice of soil type induces important variations in emissions to water f_{sw} and f_{gw} but causes very few changes in f_{air}. However, detailed soil description is time consuming and/or costly, hence not available for all vineyard situations.

Concerning the role of sprayer type in PestLCI 2.0, results of herbicides emissions are nearly not affected by the choice of weeding boom type; in contrast, the type of sprayer chosen for applications on vine canopy is the third most important driver of f_{air} variation.

4.3 Comparison to simplified emission/inventory modelling approaches

Large differences in emissions and impacts were found between the two simplified emission/inventory modelling approaches (Ecoinvent and Neto et al. (2012)) and PestLCI 2.0-based emission quantification. The definition of system boundaries is shown to have considerable influence on a pesticide's emissions quantification results (Dijkman et al. 2012; van Zelm et al. 2014). In the studies presented by Nemecek and Schnetzer (2011), Neto et al. (2012) and Petti et al. (2006), soil (in general, including agricultural soil) is considered part of the ecosphere, and all pesticide transfers to this compartment are considered emissions to the ecosphere. The PestLCI model, in contrast, considers the entire field parcel as part of the technosphere including the top 1-m soil and a 100-m air column above it (Birkved and Hauschild 2006; Dijkman et al. 2012), and models fate of chemicals within the technosphere and emissions to the ecosphere (Dijkman et al. 2013). This choice was done considering that agricultural fields are highly manipulated and controlled and therefore not "natural". Accounting for the sole emissions that cross the parcel borders is a first element limiting the quantity of emitted pesticides as modelled by PestLCI 2.0, compared to the other approaches tested. However, that is not the only cause of lower emissions and FwEtox; considering processes of evaporation, runoff and leaching, including the actual properties of the PAIs applied, canopy influence, soils and sprayers all allow for a more accurate

adjustment of estimates to the real phenomena. Degradation of PAIs and their uptake by the plants are actual processes that are not considered in the simplified emission modelling approaches tested but accounted for in PestLCI 2.0.

A "100 % emission to agricultural soil" assumption, as done in Ecoinvent, at first glance appears to be rather conservative (e.g. interception by the crop is completely neglected etc.). However, the available life cycle impact assessment (LCIA) methods (e.g. USES-LCA (van Zelm et al. 2009), CML 2002 (Guinee 2002), etc.) differ in their system boundaries and assumptions. Some of these LCIA methods model agricultural system-ecosphere transfers, and the inventory just needs to quantify the amount of PAIs emitted from the sprayer. Ecoinvent's "100 % emissions to agricultural soil" assumption is relevant in the case of use of these specific LCIA methods (Nemecek, personal communication 2014); nevertheless, site and application technique-specific conditions' influence on the emissions cannot be accounted for applying this standard Ecoinvent emission quantification approach.

In the case of use of LCIA methods that do not model the transfer from agricultural system to ecosphere and degradation processes as USEToy™, this "100 % emissions to agricultural soil" assumption might lead, as shown in the present study, to the overestimation of impacts to soil or also to the underestimation to impacts in water and air. Thus, the pesticide emission fractions need to be improved by the LCA practitioners on a case to case basis potentially taking into account dynamic issues which cannot be handled by inventory databases. This assessor-driven improvement of the pesticide emission profiles, however, is only in few (including the present case) performed. Further applying complex inventory models like PestLCI is a time- and data-demanding issue. However, neglecting e.g. crop interception, will entail overestimation of the emission fractions, and hence, application of the conservative default pesticide emission profiles applied in Ecoinvent, as well as the approach used by Neto et al. (2012), will lead to an overestimation of the potential toxicity impacts induced by application of pesticides in most crop-related LCAs. Comparing the approaches applied by Ecoinvent and Neto et al. (2012) would most likely reveal that the Ecoinvent approach is the least conservative of the two approaches due to the partial immobilization of pesticides in the soil compartment combined with the effective removal/fate processes taking place in this compartment.

It is obvious that the three compared approaches yield quite different results, which may appear peculiar. One might ask if some of the considered inventory approaches are overestimating/underestimating the pesticide emissions.

Apart from the already mentioned study by Dijkman et al. (2013), little work seems to have been done in trying to answer this question, or the consequence of the different modelling approaches on freshwater ecotoxicity impacts. The question whether the inventory approaches studied here are overestimating or underestimating emission is hard if possible to answer at all, since the perception of whether the field or parts hereof belongs to the technosphere/ecosphere and hence what pesticide flows should be regarded elementary/non-elementary flows will in accordance with Hofstetter (1998) differ from assessor to assessor and hence differ depending on the way the assessor perceives the world. Since PestLCI, in line with Hofstetter (1998), considers the field as part of the technosphere, the fate processes occurring in the field are also taking place within the technosphere. Numerous fate processes take place within the technosphere (in relation to e.g. waste water treatment, bread baking, beer brewing processes, etc.); however, the fact that the in-field fate processes are handled by a pesticide dedicated fate model and not by a chemical generic characterization model is a distinctive feature of PestLCI.

4.4 Further improvements and developments

PestLCI 2.0 could be improved by further developments in the modelling of airborne drift, which can be considerable (Jensen and Olesen 2014), but the complexity of the phenomena (Gil et al. 2008) and the lack of (generic) data are considered major obstacles for this improvement. More or less for the same reasons, pesticide metabolites are not accounted for in the present version of PestLCI 2.0. Accounting for application parameters as sprayers' speed, droplets size, temperature and relative humidity would be ideal for further refinement of the modelling of the spray mixture behaviour and fate, but these parameters are too difficult to obtain from the growers and would further entail an even more complicated inventory.

Dousset et al. (2010) found that a grass cover under vines permitted a twofold to fourfold reduction of pesticides leaching to groundwater in relation with increase of PAIs sorption in the soil thanks to organic matter content increase. This question could not be addressed here but should be addressed in the further developments of PestLCI 2.0.

High percentages of stones can be found in many vineyard soils, modifying water and solutes flow in the soil. These aspects could not be included in the present customization of PestLCI 2.0. However, improvement of the way soil texture affects macropore transport in PestLCI 2.0 is recommended as an important issue to be considered in the coming PestLCI versions.

After the end of the vineyard life, the parcel can be bound to other uses and then can be considered coming back to ecosphere. The quantity of PAIs remaining in the soil after a given period (i.e. 30 or 40 years, when the vines typically are pulled out) is information that would be useful for estimating impacts of viticulture, in case of land use change. This information would be valuable inputs for soil quality indicators and could also be applied to land use changes related to agriculture in general.

The question of impacts of pesticides on the ecosystem present in the field, which is considered here as technosphere, is a controversial question (van Zelm et al. 2014), especially because in integrated farming and organic farming, this ecosystem is considered as an ally against pests and disease and should be preserved as much as possible. However, according to ILCD (European Commission Joint Research Centre 2010), "Pesticide and fertilizer applications are no emission, but part of the product flows within the (man-managed) technosphere". Hence, the question of effects of pesticides on internal ecosystems should be addressed in a different way e.g. by accounting for reduced ecosystem services by land use change (i.e. the transition from ecosphere to technosphere) or through specific biodiversity indicators.

In organic viticulture, sulphur and copper (inorganic PAIs) are the only means available to manage respectively powdery and downy mildew and represent important quantities of applied pesticides in viticulture in general, especially sulphur. As previously mentioned, PestLCI 2.0 model is designed only for organic PAI emissions modelling. Thus, a comparison between conventional and organic viticulture or the inclusion of organically managed cases in a study cannot be dealt with solely through PestLCI 2.0. In contrast to pesticides, ILCD (European Commission Joint Research Centre 2010) points out the fact that "some inputs to soil do not leave the technosphere via leaching etc., but are accumulated in the soil. The amount/…/ applied to the field is directly inventoried as emission to agricultural soil", the latter is also the case for copper used as pesticide in viticulture (Mackie et al. 2012) that should thus be inventoried as heavy metal. Nevertheless, the primary distribution should be calculated first, especially to quantify drifted copper to ecosphere. A model similar to PestLCI is needed for emissions modelling of other inorganic pesticides. Upon release, inorganic chemicals undergo speciation (meaning that an e.g. copper emission to arable land simply cannot be modelled as an emission of e.g. Cu^{2+} but should be modelled as a set of species ($CuOH^+$, $CuCl^+$, $CuCO_3$, Cu_2^+, Cu^+, $CuSO_4$, etc.). Many of such species do not degrade as organic chemicals do, and the fate modelling of inorganic emission is typically focused on the removal of such species (via burial in sediments, leaching in soils, etc.) from the part of the ecosphere, where interaction with biological receptors may occur (i.e. the part of the ecosphere where (eco)toxicological effects may occur). Modelling the behaviour of inorganic emissions to arable land hence demands a different approach than when modelling emissions of organic chemicals. These differences are so large that in order to model inorganic pesticides

appropriately in PestLCI, a range of new sub-models for inorganic chemicals would have to be developed for PestLCI.

An additional, however important, issue is whether the overall uncertainty improvements provided by highly specific/detailed inventory approaches such as PestLCI make sense keeping in mind the considerable uncertainties related with other steps in LCA e.g. characterization of chemical emissions. We think that if any uncertainty aspect in LCA can be improved, it should be improved irrespective of whether other steps in LCA currently can or cannot match such uncertainty improvements. LCA is still developing, and chemical characterization in LCA will also at some point in time mature (and thus move beyond consensus) in terms of uncertainty.

5 Conclusions

While having been intended mainly for arable crops, the PestLCI 2.0 inventory model, due to its rather flexible framework, has here been adapted for viticulture without compromising the model framework. The PestLCI 2.0 customized version for viticulture, presented in the paper at hand, facilitates the calculations of emission loads for vertically trained vineyards with a wide range of sprayers. It further provides a considerable, though non-exhaustive, PestLCI pesticide database update of viticulture-specific PAIs, completed by the corresponding USEtox™ FwEtox CFs, and it allows taking into account cover crop effect on PAIs emissions. High variability of PAI emissions and FwEtox due to pesticides properties, spraying and environmental conditions, and comparison with simplified emission modelling approaches of pesticides PAIs emissions quantification show the interest of substance- and conditions-specific modelling with PestLCI.

Finally, some of the new PestLCI model parameters can also be used for other perennial or bush crops as long as equipment, canopy shape and PAIs stay in the range of available options.

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