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1 **Residues of currently used pesticides in soils and earthworms: a silent**
2 **threat?**

3

4 Pelosi C.^{1*}, Bertrand C.², Daniele G.³, Coeurdassier M.⁴, Benoit P.⁵, Néliu S.⁵, Lafay F.³, Bretagnolle
5 V.^{6,7}, Gaba S.^{7,8}, Vulliet, E.³, Fritsch C.⁴

6

7 ¹ INRAE, Avignon Université, UMR EMMAH, F-84000, Avignon, France - celine.pelosi@inrae.fr

8 ² Université Paris-Saclay, INRAE, AgroParisTech, UMR ECOSYS, 78026, Versailles, France –
9 colette.bertrand@inrae.fr

10 ³ UMR 5280 Institut des Sciences analytiques, CNRS, Université de Lyon, 69100, Lyon, France –
11 gaelle.daniele@isa-lyon.fr

12 ⁴ UMR 6249 Chrono-environnement CNRS - Université Bourgogne Franche-Comté Usc INRAE, 16
13 route de Gray 25030 Besançon cedex, France – clementine.fritsch@univ-fcomte.fr

14 ⁵ Université Paris-Saclay, INRAE, AgroParisTech, UMR ECOSYS, 78850, Thiverval-Grignon,
15 France – pierre.benoit@inrae.fr

16 ⁶ UMR 7372 CEBC, CNRS, Université de La Rochelle, 79360, Chizé, France –
17 vincent.bretagnolle@cebc.cnrs.fr

18 ⁷ LTSER « Zone Atelier Plaine & Val de Sèvre », Beauvoir sur Niort, 79360, France

19 ⁸ USC 1339 Centre d'Etudes Biologiques de Chizé, INRAE, F-76390 Villiers-en-Bois, France

20

21 *Corresponding author: celine.pelosi@inrae.fr

22 INRAE, Avignon Université, UMR EMMAH, F-84000, Avignon, France. 228 route de l'Aérodrome,
23 CS 40 509, 84 914 AVIGNON Cedex 9, France. Tel: +33 (0)4 32 72 22 28; Fax: +33 (0)4 32 72 22

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26

27 **Abstract**

28 Critical knowledge gaps about environmental fate and unintentional effects of currently used
29 pesticides (CUPs) hamper the understanding and mitigation of their global impacts on ecological
30 processes. We investigated the exposure of earthworms to 31 multiclass CUPs in an arable landscape
31 in France. We highlighted the presence of at least one pesticide in all soils (n=180) and 92% of
32 earthworms (n=155) both in treated crops and nontreated habitats (hedgerows, grasslands, and cereals
33 under organic farming). Mixtures of at least one insecticide, one herbicide, and one fungicide (> limit
34 of quantification) contaminated 90% of soils and 54% of earthworms at levels that could endanger
35 these nontarget beneficial soil organisms. A high risk of chronic toxicity to earthworms was found
36 (46% of samples) both in treated winter cereals and nontreated habitats considered as refuges. This
37 may alter biodiversity, hinder recovery, and impair ecosystem functions. These results provide
38 essential insights for sustainable agriculture and CUP regulation, and highlight the potential of
39 pesticides as agents of global change.

40

41 **Keywords:** soil-dwelling invertebrates, agroecology, plant protection products, risk assessment,
42 nontarget wildlife.

43

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45

46 **1. Introduction**

47 Worldwide, the diversity and quantity per hectare of synthetic pesticides used are increasing, along
48 with an increase in the area of treated surfaces (Bernhardt et al., 2017; DiBartolomeis et al., 2019;
49 Hossard et al., 2017). Global pesticide use (in tons of active ingredients) increased by 80% worldwide
50 between 1990 (2 285 881 tons) and 2017 (4 113 591 tons) (FAOSTAT, 2019; Zhang et al., 2011), and
51 the total sales of pesticides remained constant in Europe between 2011 and 2018, revealing that there
52 was no reduction in reliance on pesticides (Environmental indicator report, 2018; Eurostat, 2020;
53 FAOSTAT, 2019). Hundreds of thousands of formulated pesticides have been developed since the
54 1980s (Zhang et al., 2011), and 479 active ingredients are currently used in several thousands of
55 commercial products in the European Union (European Commission, 2020). Consequently, and
56 despite precautions to farmers to limit pesticide losses and efforts to reduce pesticide mobility within
57 the environment, their application leads to unavoidable transfer by spray drift, volatilization,
58 infiltration, and runoff from treated areas (Mottes et al., 2014). These processes potentially result in
59 the contamination of air (Bedos et al., 2002), soil (Silva et al., 2019) and water (Gilliom et al., 2007)
60 by currently used pesticides (CUPs), with serious concerns regarding their effects on the ecosystem
61 services provided by soils, water systems (Lautenbach et al., 2012) and wildlife (Brühl and Zaller,
62 2019; Geiger et al., 2010).

63 While water contamination by pesticides has been extensively studied for approximately 30 years
64 (Gilliom et al., 2007; Hallberg et al., 1987), data on the contamination of soils by CUPs *in natura* are
65 surprisingly scarce. However, the restoration or conservation of soils and their quality has been
66 recognized as a key issue, considering the fundamental role of soils in the ecosystem and the economy
67 (BIO Intelligence Service, 2014; EUR-Lex, 2006). Some recent data revealed the high occurrence of
68 mixtures of CUPs in soils of arable fields directly treated with pesticides (Chiaia-Hernandez et al.,
69 2017; Gamón et al., 2003; Hvězdová et al., 2018; Karasali et al., 2016; Marković et al., 2010; Silva et
70 al., 2019; Suszter and Ambrus, 2017; Zhao et al., 2018). However, no information is available on the
71 overall soil multiresidue contamination of farmland at the landscape scale (i.e., including both treated
72 and nontreated areas), except for neonicotinod class (e.g., Main et al., 2020). Indeed, no data can be

73 found on soil contamination by multiclass CUPs in off-field landscape elements corresponding to
74 seminatural habitats (e.g., hedgerows, wooded patches, field margins) or nontreated organic fields.
75 However, it is widely recognized that these habitats favor the presence of beneficial organisms in
76 agricultural landscapes (Bengtsson et al., 2005; Geiger et al., 2010) by playing an important role as a
77 refuge and source of recolonization following pesticide application (EFSA, 2016). Consequently,
78 when contaminated by pesticides, these nontreated habitats could act as ecological traps for organisms
79 due to a mismatch between habitat attractiveness and quality.

80 Animals living in close contact with the soil can be directly exposed to pesticides and harmed. It was
81 recently shown that the CUP concentrations in agricultural soils treated with pesticides exceeded the
82 toxicological benchmarks for earthworms or other soil invertebrates in 35% of the agricultural sites
83 studied (Vašíčková et al., 2019). Earthworms play a key beneficial role in soil structure, functioning
84 and productivity (Liu et al., 2019; van Groenigen et al., 2014) and are important prey for numerous
85 predators (King et al., 2010). Earthworm abundance has been shown to increase when pesticide use
86 decreases (Pelosi et al., 2013a) and to be lower in conventional than organic fields (Pelosi et al., 2015),
87 although it is difficult to isolate the effects of pesticides, due to biotic and abiotic factors operating at
88 the same time. However, there are no available data on the contamination of earthworms by multiclass
89 CUPs *in natura* in either treated or nontreated habitats in arable landscapes. Such data would provide
90 new insight into the pesticide bioaccumulation potential, likely unintentional effects of these
91 chemicals on earthworm populations, and the risks of transfer to their predators.

92 In this study, we investigated the level of contamination by CUPs in soils and earthworms in treated
93 and nontreated habitats of an intensive agricultural landscape. We checked whether multiclass residues
94 of CUPs might be detected in soils and earthworms, including some compounds that are assumed to be
95 weakly or moderately persistent in the environment, presenting low bioaccumulation potential and/or
96 are used in limited amounts (at a low dose rate, or only on certain crops). We hypothesized that the
97 contamination patterns of soils and earthworms would differ in the different habitats (grasslands,
98 cereal fields, and hedgerows) and according to the agricultural management (treated *vs* nontreated
99 habitats, organic *vs* conventional farming). We expected that the number and the concentrations of the
100 pesticides would be higher in habitats that were treated by CUPs than in seminatural habitats and

101 organic fields that are not directly targeted by pesticides. Based on the available data on the predicted
102 environmental concentrations of pesticides in soils (PEC_s provided in risk assessment documents
103 according to the European regulation) and toxic thresholds for earthworm reproduction (for each
104 pesticide separately, and using a mixture approach based on concentration addition), we also assessed
105 the risks to earthworms.

106

107 **2. Methods**

108 **2.1. Sampling area and design**

109 The sampling of soils and earthworms was conducted in Spring 2016 in the Long-Term Socio-
110 Ecological Site Zone Atelier Plaine & Val de Sèvre (ZA-PVS (Bretagnolle et al., 2018);
111 <http://www.za.plainevalsevre.cnrs.fr/>). Sixty landscapes of 1 km² were selected in which soil and
112 earthworms were sampled in an arable field sown with winter cereals, a grassland and a hedgerow or
113 woody patch edge (as close as possible to the cereal field), for a total of 180 sampling site locations
114 (Table 1). Among the 52 cereal fields where earthworms were sampled, 44, 6, and 2 were sown with
115 winter wheat, winter barley, and einkorn, respectively. The farming practices in the organic cereal
116 fields and grasslands respected the rules of the AB France label and were under organic farming for at
117 least 3 years at the time of sampling. A total of 180 soils and 155 earthworms were therefore analyzed
118 to determine pesticide concentrations (Table 1).

119

120 **2.2. Collection of soils and earthworms**

121 In each plot, regardless of the size of the sampled habitats (i.e., winter cereal fields, grasslands,
122 hedgerows), three subsamples (0–5 cm depth; Amelung et al., 2007; de Geronimo et al., 2015) were
123 taken using a 5 cm Ø soil auger. They were then combined to obtain one composite sample per site.
124 The depth of 5 cm was chosen because the soils in the sampling area were shallow and rocky,
125 sometimes not allowing to sample at more than 5 cm depth. Moreover, the studied earthworm *A.*
126 *chlorotica* is an endo-epigeic species that is commonly found in the top 5 cm of the soil (Pelosi et al.,
127 2013a; Le Couteulx et al., 2015). The soils were frozen at -20°C before being analyzed.

128 Soil properties were measured at the Laboratoire d'Analyse des Sols of the Institut National de la
129 Recherche Agronomique (Arras, France), which benefits from the COFRAC (French accreditation
130 committee) accreditation of its analytical quality regarding soil characteristics. Briefly, soils were
131 dried at room temperature and then disaggregated and homogenized before being sieved at 2 mm. The
132 following soil characteristics were measured: pH (by water suspension), organic matter and nitrogen
133 contents (by dry combustion, in g kg^{-1}), grain size distribution (clay $< 2 \mu\text{m}$, silt 2-20 μm , and sand $>$
134 20 μm , in g kg^{-1}), total calcium carbonate CaCO_3 (in g kg^{-1}), and total phosphorus P_2O_5 (by ICP-MS
135 spectrometry, in g kg^{-1}).

136 We focused on the endo-epigeic earthworm species *Allolobophora chlorotica* which is well
137 represented in the different sampled landscape habitats in the ZA PVS. Because pesticides generally
138 accumulate at the soil surface, species living in contact with the soil surface will potentially be more
139 strongly affected than those living deeper (Pelosi et al., 2013a). Regardless of the size of the sampled
140 habitats (i.e., winter cereal fields, grasslands, hedgerows), earthworms were searched for 15-30
141 minutes at each site location by superficially digging the soil, allowing to find between 0 and 10 *A.*
142 *chlorotica* adult individuals. In 25 out of the 180 sampling site locations, *A. chlorotica* could not be
143 found. Before being weighed and frozen at -80°C , earthworms were individually placed in petri dishes
144 on damp filter paper for 48 h to void their gut contents.

145

146 **2.3. Analytics for residues of pesticides**

147 The analyzed pesticides (Table S1) were selected based on analytical capabilities as well as their
148 frequency and amount of application over the sampling area recorded in surveys of farmers over the
149 last 5 years before sampling. Thirty-one pesticides (9 insecticides, 10 fungicides, and 12 herbicides,
150 see Tables 2 and 3) were studied, 29 of which were still registered and used at the time of sampling,
151 while 2 were recently banned pesticides (acetochlor and bifenthrine, banned in 2013). They were all
152 referred to as Currently Used Pesticides (CUPs) in this study. For analytical reasons or because they
153 were applied after the sampling date, some pesticides had been applied over the sampling area but
154 were not measured in this study such as e.g., glyphosate, prothioconazole, metaldehyde, florasulam,

155 pinoxaden, picolinafen, or isoproturon. We also voluntarily limited the number of active substances in
156 the analyses to keep low limits of detection (LOD) and quantification (LOQ).

157 The recommended dose (RD) of each active substance was calculated considering the commercial
158 formulations currently used on cereal crops, penetration of 5 cm depth, and a soil bulk density of 1.5
159 (EFSA, 2017).

160 An analytical multiresidue method has been implemented and validated (Daniele et al., 2018; Daniele
161 et al., in press) to measure 31 pesticides in soils and earthworms. As pesticides are sensitive to
162 temperature, soil samples were air dried at room temperature in the dark during one night. The soil
163 was sieved at 250 μm before extraction. The LOD and LOQ are provided in Table S1. Because LOD
164 and LOQ values were different for each compound, we chose to always consider what was the LOQ
165 for saying positive/negative. Briefly, a modified QuEChERS extraction approach was implemented for
166 individual earthworms (aliquots of 250-mg wet weight, i.e., between 1 and 3 earthworm individuals)
167 using water (6 ml), heptane (3 ml) and two successive extractions were performed with acetonitrile (5
168 ml), citrate salt and a PSA/C18 clean-up step, followed by liquid chromatography coupled to tandem
169 mass spectrometry (LC-MS/MS). For soil analysis, QuEChERS extraction (citrate salt) was conducted
170 for 2.5 g of dried and sieved soils using water (6 ml, containing 0.1 M EDTA), and two successive
171 extractions were performed with 5 ml of acetonitrile in the presence of citrate buffer, followed by
172 dispersive solid-phase extraction with a PSA/C18 phase. The extracts were analyzed by using LC-
173 MS/MS. The instrumental performance and eventual carry-over have been controlled regularly by
174 injecting quality control and analytical blank samples, respectively.

175

176 **2.4. Risk assessment**

177 The predicted environmental concentrations in soils (PECs) and acute (LC50) or chronic (NOEC
178 reproduction) toxicity thresholds for earthworms (*Eisenia fetida*) were collected from evaluation
179 reports provided according to European Directives regarding the registration of plant protection
180 products under the authority of the “Health & consumer protection directorate-general of the European
181 Commission” and the “European Food Safety Authority” (European Commission, 2003; European
182 Parliament and Council of the European Union, 2009). Toxicity values were checked and updated if

183 necessary based on information from the “Pesticide Properties DataBase”
184 (<http://sitem.herts.ac.uk/aeru/ppdb/index.htm>). Alternatively, when the parameters of interest were not
185 provided in these sources, other reports of risk assessments (e.g., postregistration, authority of national
186 agencies) and scientific publications were searched.

187 The PECs are concentrations expected in agricultural soils under worst case conditions in scenarios of
188 authorized commercial use for each given compound, that are obtained from modelling and/or
189 measured concentrations in trials. The measured concentrations in soils (MECs) are thus supposed to
190 be equal to or lower than the maximum PECs. The values of PECs are used in risk assessment
191 procedures to calculate the toxicity/exposure ratio, which is a crucial endpoint to determine whether a
192 risk to organisms can arise from the use of the compound under allowed practices at recommended
193 doses, and therefore determine the marketing authorization. We here used the PECs values provided in
194 registration documents calculated for the same crop that studied in our dataset (i.e., wheat) when
195 available or similar application scheme on other crops (e.g., general case cereals) at recommended
196 application rates. In order to provide quantitative data about the general patterns of contamination with
197 regards to expected levels in the environment, we compared MECs to PECs for each compound.

198 Indeed, to get further insights into the ecotoxicological significance and the efficiency of risk
199 assessment procedure, comparing MECs to PECs is a way to highlight whether levels of residues in
200 soils occur at « trace levels » both in treated and nontreated plots with regards to potential risk and
201 allowed practices. Since no data about time of application and detailed practices in each plot were
202 available, several PECs values related to « worst cases » and used to calculate toxicity ratio for soil
203 fauna, such as PECs initial after treatment, long term PECs and maximum PECs were considered. The
204 fact that MECs can be higher than PECs in soils where compounds are used under normal scenario or
205 where a compound might not have been applied at all is an important result to enlight the spatial
206 patterns of pesticide contamination in terrestrial environment.

207 A single-pesticide approach was applied first using the toxicity/exposure ratio for earthworms
208 ($TER_{\text{earthworm}}$). This approach follows the risk assessment method for pesticide regulation defined by
209 European legislation and has been used in recent scientific studies (e.g., Vašíčková et al., 2019). The
210 value of $TER_{\text{earthworm}}$ was calculated for each soil sample as the ratio between the values of LC50 or

211 NOEC divided by the measured soil concentrations above the limits of detection for each CUP
212 individually. When thresholds were provided as “greater than” values, the given benchmarks were
213 used in the calculations. The risk was considered negligible when the TERearthworm values were
214 above a trigger limit of 10 for acute toxicity and of 5 for chronic toxicity following European
215 regulations. As an example, considering epoxiconazole, the TER for acute toxicity and chronic
216 toxicity were calculated for each sample as a LC50 of 62500 ng g⁻¹ and a NOEC of 84 ng g⁻¹,
217 respectively, divided by epoxiconazole concentration in soil. In case the acute toxicity TER was higher
218 than 10, the risk was considered negligible, which was the case for all samples. In case the chronic
219 toxicity TER was higher than 5, the risk is considered negligible, which was not the case for 52 soil
220 samples in which the calculated TER value was under or equal to this trigger of 5.

221 Then, a mixture approach was applied to assess the risks related to the presence of several residues in
222 the samples. The risk quotient (RQ), as primarily used by the US EPA (2017), was computed for each
223 single CUP as the ratio between the measured environmental concentrations (when above the limits of
224 detection) divided by predicted no effect concentration (PNEC) for each soil sample. The PNEC
225 values were computed as the most susceptible endpoint, i.e., the NOEC or, if not available, the LC50,
226 divided by the recommended assessment factors (AF). Assessment factors were derived from the
227 instructions of the Environmental Risk Assessment Guidance (European Commission, 2003) using
228 1000 for the LC50 (AF for short-term toxicity test) and 10 for the NOEC (AF for long-term toxicity
229 tests; since we focused on earthworms, the application of the criteria related to the number of trophic
230 levels of the targets was not performed).

231 Finally, an additional approach was applied as recommended to assess the multiple toxicity of several
232 pesticides in the guidelines of the European Food Safety Authority for risk assessment for birds and
233 mammals (EFSA, 2019). Despite some drawbacks of such a use of the concentration addition concept
234 (e.g., synergistic effects are not considered), no alternative reliable and validated method is available
235 or routinely applied. The addition concept is broadly accepted by authorities around the world and
236 used in scientific publications (e.g., Vašíčková et al., 2019). The individual RQ values for every CUP
237 were summed ($\sum RQ$) for each soil sample. Finally, the $\sum RQ$ values were classified into four

238 categories: high risk ($\sum RQ \geq 1$), medium risk ($0.1 \leq \sum RQ < 1$), low risk ($0.01 \leq \sum RQ < 0.1$) and
239 negligible risk ($\sum RQ \leq 0.01$) (US EPA, 2017; Vašíčková et al., 2019).

240

241 **2.5. Statistics**

242 When a pesticide was not detected in a sample (value < LOD), the concentration value was set at 0
243 when necessary for statistical method application. When a pesticide was detected at a level below the
244 LOQ but above the LOD, the LOD value was attributed.

245 ANOVA (or the Kruskal-Wallis test, when assumptions regarding the normality and homoscedasticity
246 of variances were not respected) was used to assess the differences in earthworm and soil pesticide
247 variables (i.e., number of pesticides and concentrations) between the three habitats (i.e., cereal fields,
248 grasslands, hedgerows). The t-test (or the Wilcoxon test when assumptions regarding the normality
249 and homoscedasticity of variances were not respected) was used to assess the differences in earthworm
250 and soil pesticide variables between the two modalities of pesticide use (treated/nontreated). For the
251 differences between conventional and organic fields and grasslands (T-test or Wilcoxon test), the data
252 from the hedgerows (or woody patches) were removed from the dataset.

253 Multivariate conditional inference trees were used to cluster soils and earthworms according to the
254 relationships between the patterns of soil or earthworm contamination (response variables:
255 concentrations of CUPs) and four explanatory variables: type of habitat, treated/nontreated by
256 pesticides, organic matter and clay contents. The last two parameters were chosen as they influence
257 the most the fate and accumulation of pesticides in the studied matrices. In addition, they were not
258 correlated with each other. pH was not included in the MRT analyses since 83% of the values were
259 between 8 and 8.5 and were thus not potentially discriminant in the analysis. The number of variables
260 in the MRT analyses was deliberately kept small to maintain sufficient statistical power. Pesticides
261 that had never been detected were removed from MRT analysis because they were not discriminating.
262 Spearman correlation tests were used to test the relationships between the number of years since the
263 switch to organic farming (in cereal fields and grasslands) and the numbers or concentrations of CUPs
264 for earthworms and soils.

265 All statistical analyses were performed in RStudio version 3.3.2 using the following packages: partykit
266 (Hothorn and Zeileis, 2015), pgirmess (Giraudoux et al., 2017), and car (Fox and Weisberg, 2019) for
267 the other analysis.

268

269 **3. Results**

270 **3.1. CUPs in soils and earthworms**

271 Among the 31 CUPs analyzed, 27 were detected in soils (Table 2). All the soils contained at least one
272 CUP (n = 180), 83% exhibited five CUPs or more, and 38% exhibited ten or more (Figure 1). The
273 herbicide diflufenican, the insecticide imidacloprid, and the fungicides boscalid and epoxiconazole
274 were found in 90%, 89%, 85%, and 79% of the soils, respectively (Table 2). The most common
275 mixture consisted of an insecticide (imidacloprid), an herbicide (diflufenican), and a fungicide
276 (boscalid (74% of the soils), epoxiconazole (71%), or prochloraz (48%)). Some pesticides were found
277 at relatively low concentrations (<10 ng g⁻¹) in soils, but others, such as diflufenican, boscalid, and the
278 herbicide pendimethalin, reached concentrations >100 ng g⁻¹, or >500 ng g⁻¹ (Table 2, Figure 2a). For
279 instance, boscalid was measured at a concentration of 1212 ng g⁻¹ in a cereal field under conventional
280 farming, corresponding to 2.3 times the recommended dose (RD) (Table S1). For diflufenican (1361
281 ng g⁻¹, representing 4.9 times the RD) and prochloraz (485 ng g⁻¹, or 0.7 times the RD), the highest
282 concentrations were found in the same cereal field (Table 2, Table S1). The four nondetected
283 pesticides were the herbicide cycloxydim and the pyrethroid insecticides bifenthrin, deltamethrin, and
284 lambda-cyhalothrin, which was consistent with their limited use on the sampled crops and their low
285 persistence in soils (Table S1).

286 In earthworms, 18 CUPs were detected among the 31 CUPs analyzed (Table 3). The mean number of
287 CUPs per earthworm (3.5 ± 2.2 pesticides per individual, n = 155) was lower than that in soils ($8.5 \pm$
288 4.1 pesticides per soil sample, n = 180) (Figure 1), but higher concentrations were measured in
289 earthworms for some pesticides, such as diflufenican and imidacloprid (Figure 2, Tables 2 and 3). Up
290 to 11 pesticides were found in one earthworm sampled in a winter wheat field (Figure 1). Ninety-two
291 percent of earthworms contained at least one of the pesticides, and 34% (n = 52) exhibited five

292 pesticides or more (Figure 1). Overall, imidacloprid was the most frequently detected CUP regardless
293 of the habitat (cereal fields, hedgerows, grasslands) and farming system (conventional *vs* organic
294 farming), with 79% of individuals being positive for imidacloprid (Table 2). This insecticide also
295 showed the highest frequency of high concentrations, with 43% of the earthworms presenting
296 imidacloprid concentrations $>100 \text{ ng g}^{-1}$ and 8.4% $>500 \text{ ng g}^{-1}$ (Figure 2b). The mean concentration of
297 imidacloprid in earthworms was the highest among all the CUPs analyzed in the three landscape
298 habitats, followed by diflufenican in cereal fields and hedgerows and epoxiconazole in grasslands
299 (Table 4). The highest concentrations in a single individual were found for two herbicides,
300 pendimethalin and diflufenican, one fungicide, prochloraz, and one insecticide, imidacloprid (Table 3).
301 The most frequent mixture in earthworms was the same as that in soils, i.e., imidacloprid, diflufenican
302 and one of two fungicides, epoxiconazole (33% of the earthworms) or cyproconazole (27%).

303

304 **3.2. Patterns of contamination according to habitats and agricultural management**

305 The pesticide contamination patterns of the soils differed first according to habitat type (Figure 3a).
306 The soil contamination profiles associated with cereals were characterized by a greater number of
307 pesticides, relatively high concentrations of diflufenican, imidacloprid, boscalid, epoxiconazole,
308 prochloraz and pendimethalin, and a high occurrence (number of samples in which the pesticide was
309 detected) of cyproconazole compared to those in soils from grasslands and hedgerows (Figure 3a). In
310 addition, a greater number of herbicides, fungicides, or insecticides was found in soils from cereal
311 fields than in soils from other habitats (Table 4). The occurrence and concentration of the five most
312 frequent pesticides in soils (Table 2) were higher in cereal fields than in hedgerows and grasslands
313 (Table 4). The diflufenican, imidacloprid, boscalid, epoxiconazole, and prochloraz concentrations
314 were 18, 5, 15, 6, and 45 times lower, respectively, in hedgerows and grasslands than in cereal fields.
315 Regardless of the class of CUPs, the soil of fields treated with pesticides exhibited a greater number of
316 pesticides than those of nontreated habitats (i.e., hedgerows, organic cereal fields or grasslands, and
317 permanent grasslands) (Table 4), although the factor “treated or nontreated” did not shape the patterns
318 of soil contamination (Figure 3a). The difference was less noticeable for insecticides than for the other
319 pesticides, as 67%, 56%, and 29% greater number of herbicides, fungicides, and insecticides (mean

320 number of pesticides per sample), respectively, were found in soils from treated than nontreated
321 habitats (Table 4, Table S2). Moreover, similar concentrations of boscalid were found in
322 treated/nontreated habitats as well as in organic and conventional fields. Among the 93 soil samples
323 collected in the nontreated habitats, 83% contained more than 3 pesticides. The comparison of soil
324 contamination between conventional and organic farming revealed that the number of CUPs found in
325 soils from conventional fields was 63% higher for insecticides, 89% higher for fungicides, and 68%
326 higher for herbicides (Table 4, Table S2). However, 83% of the soils under organic farming exhibited
327 three pesticides or more; 72% contained imidacloprid (from 0.4 to 7.7 ng g⁻¹) and 61% were
328 contaminated by diflufenican (from 0.1 to 4.2 ng g⁻¹). On average, 6 pesticides per soil were found in
329 cereal fields under organic farming, with three soils (i.e., 43% of the samples) containing nine
330 pesticides or more. In grasslands under organic farming, 5 pesticides per soil were detected on
331 average, and one of the samples was contaminated by 14 pesticides.

332 Similar to the results for soils, earthworm contamination profiles differed according to habitat type and
333 they were not segregated by the treated/nontreated factor (Figure 3b). The profiles associated with
334 cereals were characterized by a greater number of pesticides per earthworm than those in hedgerows
335 and grasslands. As found in soils, greater numbers of total CUPs (all classes), herbicides, or fungicides
336 were found in earthworms from cereal fields than in the other two habitats (Table 4). However, the
337 mean number of insecticides per individual was similar in cereal fields and hedgerows. It was also not
338 significantly different between treated and nontreated habitats (Table 4), with on average 1 (±0.8) and
339 1 (±0.6) pesticides per earthworm, respectively. The number of CUPs per earthworm was greater in
340 conventional fields than in organic fields for all classes of pesticides (Table 4; Table S2). However,
341 among the seven earthworms from organic cereal fields, four contained between 2 and 6 pesticides,
342 and five exhibited imidacloprid concentrations ranging from 27.2 to 110.0 ng g⁻¹ (mean 47.3 ng g⁻¹).
343 Similarly, among the ten earthworms from organic grasslands, two contained 43.3 and 102.0 ng g⁻¹
344 imidacloprid. The other CUPs presented very low concentrations (<10 ng g⁻¹) in the earthworms
345 sampled in organic cereal fields and organic grasslands.

346 Earthworms from cereal fields were characterized by relatively high concentrations of imidacloprid,
347 diflufenican, and cyproconazole and high occurrences of epoxiconazole, prochloraz and pyroxsulam

348 compared to those in soils from grasslands and hedgerows (Figure 3b). For instance, the
349 concentrations of imidacloprid, diflufenican, and epoxiconazole were between 3 (epoxiconazole, in
350 cereal fields *vs* grasslands) and 72 times (diflufenican, in cereal fields *vs* grasslands) higher in
351 earthworms sampled in cereal fields (Table 4). These results highlight the considerable weight of
352 diflufenican, imidacloprid, boscalid, epoxiconazole, prochloraz, and cyproconazole in both soil and
353 earthworm contamination patterns. While the load of pendimethalin also shaped the profiles of CUPs
354 in soils from cereal plots and grasslands, the presence of pyroxsulam was discriminant for CUP
355 profiles in earthworms. The pesticides that drove the patterns were thus not necessarily the most
356 frequent ones (Tables 2, 3 and Figure 3). Finally, we tested the correlations between the number of
357 years since the switch to organic farming and the number of CUPs or the concentrations of pesticides
358 in earthworms and soils, but no significant relationships (Spearman correlation) were found.

359

360 **3.3. Risk to earthworms exposed to a single CUP or mixture**

361 The predicted environmental concentrations in soils (PEC_s) were exceeded for 5 to 11 pesticides (in 14
362 to 170 soils, or 8 to 94% of samples, respectively) depending on the considered type of PEC_s (e.g., the
363 initial concentration after treatment, or the long-term, plateau or maximum concentration; Table 5,
364 Table S3). The main pesticides reaching levels higher than the PEC_s were boscalid, cyproconazole,
365 epoxiconazole, prochloraz (fungicides, up to 5 times higher than the initial PEC_s), diflufenican,
366 pyroxsulam (herbicides, up to 4 times higher than the initial PEC_s), and imidacloprid (insecticide, 1.03
367 times higher than the initial PEC_s). The initial PEC_s were exceeded for 7 pesticides in 22% of samples,
368 mostly in soils from conventional cereal plots but also in nontreated soils from hedgerows in 10% of
369 cases. The maximum PEC_s were exceeded for boscalid, cyproconazole, epoxiconazole, diflufenican
370 and pyroxsulam in 18% of soil samples (n = 32) collected in conventional cereal plots and in
371 hedgerows (n = 3) (Table 5).

372 Considering the single pesticide approach based on the toxicity/exposure ratio for earthworms
373 (TER_{earthworm}), no acute risk of the measured concentrations in soils was found (Table 5). However, a
374 risk of chronic toxicity was indicated for 4 pesticides, boscalid, cyproconazole, epoxiconazole or
375 imidacloprid, in 42% of soils (Table 5, Table S3). Seventy-six percent of these soils were sampled in

376 conventional cereal plots, while 12% came from hedgerows in which epoxiconazole or imidacloprid
377 exceeded toxic levels for earthworm reproduction. Moreover, epoxiconazole was found to potentially
378 alter earthworm reproduction in several grasslands (n = 8) and one organic cereal plot.
379 Regarding mixture toxicity, a high risk was found in 46% of the soil samples (Table 6). Considering
380 that the trigger value for the high risk level was set as $\sum RQ \geq 1$, high $\sum RQ$ values were found, up to
381 38 in cultivated soils and 21 in seminatural habitats (Table 6). The pesticide mixtures in soils posed a
382 negligible or low risk in only 22% of soils, and no conventional cereal plot presented a low or
383 negligible risk (Table 6). Even nontreated soils from organic cereal fields and hedgerows displayed a
384 high risk in 3 and 22 samples, respectively, representing 43% and 37% of the organic cereal field and
385 hedgerow samples. A high risk in soils from grasslands occurred only under conventional farming (5%
386 of all the soils sampled, or 19% of the grassland soils). Grasslands under organic farming were the
387 only habitat where the CUP mixture in the soils was not classified as high risk. The pesticides in the
388 mixture that mostly contributed to the risk were the same as those under the TER approach (boscalid,
389 cyproconazole, epoxiconazole and imidacloprid), in addition to propiconazole and pyraclostrobin.

390

391 **4. Discussion**

392 **4.1. CUPs in soils**

393 **4.1.1. Ubiquity of contamination in soils**

394 The first result of great importance in this study was the wide contamination of soils at the scale of an
395 agricultural landscape, as 100% of the soils sampled in conventional fields, in plots managed under
396 organic farming and in off-field habitats contained CUPs. Overall, although the levels of most of the
397 pesticides in our soils were within the ranges reported in recent studies (e.g., Chiaia-Hernandez et al.,
398 2017; Karasali et al., 2016; Suszter and Ambrus, 2017), we measured relatively high occurrences and
399 concentrations of CUPs, mainly for diflufenican, imidacloprid, boscalid, and epoxiconazole (i.e., all
400 were found in more than 80% of the samples, at up to 1361 ng g⁻¹). Among the most notable
401 differences, Silva et al. (2019) reported a maximum value of 410 ng g⁻¹ for boscalid while we
402 measured a concentration up to 1211 ng g⁻¹ in soil from a cereal field. Similarly, these authors found

403 that imidacloprid was present in 7% of the examined EU topsoil samples, based on a limit of
404 quantification of 10 ng g⁻¹, with a maximum content of 60 ng g⁻¹, while we found imidacloprid in 90%
405 of soils (or 26% of soils when considering concentrations above 10 ng g⁻¹), and at concentrations as
406 high as 160 ng g⁻¹. Lower concentrations of imidacloprid (between <0.09 and 10.7 ng g⁻¹) and
407 thiamethoxam (between <0.02 and 1.5 ng g⁻¹) have also been measured in arable soils in England,
408 where neonicotinoids have been used as seed dressings (Jones et al., 2014). Numerous nonexclusive
409 factors related to environmental conditions, type of crop studied, agronomic practices and pesticide
410 properties as well as sampling time (e.g., date since last applications), sampling strategies and
411 analytical methods (e.g., limits of detection and quantification) may drive the differences observed
412 between the present results and the previous studies (Bonmatin et al., 2015). Further investigations
413 will be required to identify and disentangle these factors but the levels of CUPs found here in soils
414 from treated and nontreated habitats suggest higher persistence and/or inputs than expected.

415

416 **4.1.2. Mixture of pesticides in soils**

417 A striking result of our study was the contamination of soils by a mixture of multiclass CUPs with
418 different chemical characteristics, modes of action and targets, since a mixture consisting of at least
419 one herbicide, fungicide and insecticide was found in 90% of the soils. However, most of these CUPs
420 are assumed to be weakly or moderately persistent in the environment. It is worth pinpointing that
421 only 31 pesticides were analyzed while about 60 active ingredients were found to be applied in the
422 studied area. In cereal fields, we detected an average of 11 pesticides in the soils (i.e., 35% of the
423 analyzed pesticide), which was higher than previous observations (e.g., 10–15 pesticides per soil in
424 treated fields corresponding to 10-16% of the analyzed pesticide in Chiaia-Hernandez et al., 2017).
425 Moreover, the percentage of soils containing at least 5 pesticides was 83%, which was higher than that
426 previously reported for soils collected in arable lands (51%) (Hvězdová et al., 2018). This is even
427 more striking when considering that we also sampled soils in nontreated habitats. As mentioned in the
428 previous sub-section, numerous factors may explain these differences between studies. We detected
429 diflufenican, imidacloprid, boscalid, and epoxiconazole most frequently (i.e., in >80% of soils), which
430 was consistent with previous findings in farmland soils indicating that epoxiconazole and diflufenican

431 or boscalid showed the highest occurrence (Hvězdová et al., 2018; Silva et al., 2019). Neonicotinoids
432 (notably imidacloprid) have rarely been measured in arable soils, although these compounds are of
433 high environmental concern regarding their potential negative impacts on biodiversity and ecosystem
434 functioning worldwide (van der Sluijs et al., 2015). Imidacloprid has also been detected at a high
435 frequency in vegetable crop fields in Jordan (Kailani et al., 2019) or in France, where it was detected
436 in 91% of sampled soils (Bonmatin et al., 2015). In this last study, 97% of soils seeded with treated
437 seeds 1 or 2 years before sampling were still contaminated by imidacloprid, a neonicotinoid that
438 potentially exhibits long persistence in the environment (Jones et al., 2014; van der Sluijs et al., 2015).
439

440 **4.1.3. CUPs contaminate soils in both treated and nontreated habitats**

441 One of our main findings was the ubiquity of the CUPs in all habitats of the agricultural landscape,
442 regardless of whether they had been treated with pesticides. Although the concentrations and the
443 number of pesticides were higher in soils sampled in habitats that directly received pesticides, we
444 identified different mixtures of CUPs in nontreated off-field habitats; for instance, an average of 6
445 pesticides per soil was found in organic cereal fields. To our knowledge, this is the first time that data
446 showing the wide contamination of nontreated habitats have been reported, since previous studies
447 dealing with pesticides in mixture mainly focused on treated cropped fields (e.g., Chiaia-Hernandez et
448 al., 2017; Hvězdová et al., 2018; Silva et al., 2019). The rare studies considering off-field or
449 nontreated areas dealt only with neonicotinoids, and Bonmatin et al (2005) did not detect any residues
450 of imidacloprid (limit of quantification at 1 ng g^{-1}) in French organic soils. However, repeated and
451 massive use of neonicotinoids in arable landscape may have modified this pattern with time course.
452 The processes explaining the contamination of nontreated habitats measured in our study could
453 include horizontal transfer via air and water from treated to nontreated habitats along with residues of
454 the applied chemicals (Navarro et al., 2007). Jones et al. (2014) detected the neonicotinoids
455 clothianidin, thiamethoxam, and imidacloprid in edges or several fields where these chemicals had not
456 been used in the three previous years and suggested that this may have been due to applications in
457 surrounding fields and dust drift. Similarly, pendimethalin and imidacloprid were found in plots where
458 they were not applied by farmers, which might be partly due to CUP treatments applied in the

459 surrounding fields (Chiaia-Hernandez et al., 2017). Overall, our results suggest that habitat shaped the
460 profiles of contamination more than farming practices (conventional versus organic farming), which
461 implies that the local beneficial effects of organic farming are dampened because of neighboring
462 inputs from large surfaces treated with CUPs.

463

464 **4.1.4. Risk assessment using PECs**

465 The predicted environmental concentrations in soils (PECs) calculated within the framework of
466 environmental risk assessment methodology in Europe are key criteria for determining whether soil
467 contamination after treatments poses a risk to the soil fauna or not. We here considered « worst case »
468 values since we provided comparisons to initial PECs and maximum PECs, or plateau/long term PEC
469 for compounds that are not supposed to accumulate in soils. This means that even if the soil sampled
470 had been treated the within hours before collection, the MECs should be at maximum equal or lower
471 than the maximum or initial PECs values. As a consequence, according to the marketing authorization
472 of the products, the MECs in plots submitted to applications whatever the time of application and in
473 nontargeted plots should be under the maximum PECs value.

474 We showed that the measured concentrations in soils exceeded the initial PEC_s by factors of 1.03 to
475 5.08 for several herbicides, fungicides and insecticides, even in nontreated habitats, raising two main
476 issues. First, this leads to questions regarding the relevance of the laboratory testing and modeling
477 approaches that are used for regulation to assess degradation and accumulation and to predict the
478 environmental levels of pesticides only in treated plots. Additionally, local environmental conditions
479 influence transfer, bioavailability and persistence and, thus, may alter the fate of pesticides (Navarro et
480 al., 2007), which is not considered in PEC calculation. At the landscape scale, pesticides can be
481 applied repeatedly in a mosaic of fields, leading to the contamination of neighboring habitats by drift
482 or volatilization and run-off. Second, the efficiency of postregistration survey methods needs to be
483 reconsidered (Marković et al., 2010) since residues in soils are rarely considered.

484

485 **4.2. Mixture of multiclass CUPs over the landscape, a threat to earthworms**

486 Except from insects and especially bees, no data are currently available regarding the accumulation of
487 multiclass CUPs in nontarget fauna or regarding the risk to wildlife arising from soil pesticide
488 mixtures under realistic field conditions. We showed that soil contamination by CUPs led to the
489 accumulation of a mixture of pesticides in 92% of the earthworms sampled. High concentrations of
490 several CUPs were measured in some earthworms, with the residues of diflufenican, prochloraz or
491 pendimethalin exceeding 1000 ng g⁻¹ in 6 individuals. Overall, the ability of CUPs to bioaccumulate in
492 soil organisms remains under question. CUPs are commonly considered to show low to moderate
493 bioaccumulation compared to organochlorine pesticides, which were prohibited several years ago, but
494 empirical evidence (i.e., the measurement of residues in free-living organisms) is lacking. For the
495 earthworm *Eisenia andrei* exposed to field-contaminated soils in laboratory experiments,
496 bioaccumulation was observed only for pendimethalin in one of 4 tested soils but not for
497 epoxiconazole and prochloraz (Neuwirthová et al., 2019). In our study, 3 pesticides that are among the
498 most frequently detected pesticides in *Allolobophora chlorotica*, diflufenican, imidacloprid, and
499 epoxiconazole, exhibited higher concentrations in earthworms than in soils. Neonicotinoids (notably
500 imidacloprid) have rarely been measured in wildlife apart from pollinators, although these compounds
501 are of high environmental concern regarding their potential negative impacts on biodiversity and
502 ecosystem functioning worldwide (van der Sluijs et al., 2015). The only study concerning CUP
503 accumulation in free-living earthworms reported levels of some neonicotinoids in a few individuals
504 sampled opportunistically in two soya bean plots. The authors have detected imidacloprid at
505 concentrations of 25 and 23 ng g⁻¹, and total neonicotinoid concentrations reached 54 and 279 ng g⁻¹,
506 which support our findings about the ability of soil organisms to be exposed to and accumulate
507 neonicotinoids (Douglas et al., 2015). These results along with those reported in our study attest to the
508 bioaccumulation potential of some CUPs, at least under field conditions, suggesting a need for much
509 more field monitoring to complement lab or modeling assessment. As earthworms are the main or
510 occasional prey of numerous wildlife species, the diverse mixture of pesticides that we found in their
511 tissues gives rise to the question of whether they could play a key role as vectors of pesticides in food
512 webs and, thus, contribute to endanger their predators.

513 Our results emphasized that several single pesticides are present in soils at levels above toxic
514 thresholds for nontarget soil organisms and may therefore present a risk to earthworms. Moreover, in
515 the consideration of potential mixture toxicity, we calculated a high risk for almost half of 180 the
516 soils sampled, including organic fields, grasslands and hedgerows. This was in line with studies
517 revealing negative impacts of pesticides used in cropping systems on earthworm populations and
518 communities (Pelosi et al., 2013a, 2015; Pfiffner and Mäder, 1998). This also reinforced current
519 questions about the relevance of risk assessment procedures to biodiversity (Brühl and Zaller, 2019;
520 Wintermantel et al., 2020). Furthermore, this alarming level of risk over a large extent and various
521 landscape patches is likely to be underestimated since we analyzed only 31 pesticides, and additional
522 CUPs are used and can occur in soils, with potential synergistic deleterious effects. Moreover, the
523 earthworm species *Eisenia fetida* used in risk assessment procedures has been shown to be less
524 sensitive to pesticides than other earthworm species found in cultivated fields (Pelosi et al., 2013b;
525 Tejada et al., 2011), which can underestimate the calculated risks. Finally, PEC values and chronic
526 toxic thresholds were not available for the full set of pesticides studied, which may lower the risk
527 evaluations for several compounds. Neither the toxic threshold related to the long-term exposure of
528 earthworms to similar mixtures of several CUPs nor reference values relating CUP residues in
529 earthworm tissues to toxicological endpoints were available to further assess the potential risk to soil
530 organisms at the individual, population and community levels.

531 The fact that levels of CUPs in fields under conventional farming never presented low or negligible
532 risk but high risk to earthworms in 91% of soils seriously questions the sustainability of chemical
533 mainstream agriculture. Moreover, within agricultural landscapes, nontreated habitats such as organic
534 fields, hedgerows or permanent grasslands are assumed to promote biodiversity (EFSA, 2016;
535 Nienstedt et al., 2012), but our results give rise to the question of whether they could act as ecological
536 traps and harm animals that live there by exposing them to pesticide mixtures at relatively high
537 concentrations. The contamination of these “off-field” habitats by pesticides could affect the resilience
538 of agrosystems at the landscape scale by preventing any possibility of these areas to act as shelters and
539 sources for recolonization. It has been emphasized that the use of neonicotinoids hinders the
540 maintenance of biodiversity and the ecological functions and services the organisms perform (van der

541 Sluijs et al., 2015). Our results regarding off-field habitat contamination by CUPs bolstered this
542 conclusion and indicated that its application should also be broadened to several other pesticides,
543 notably fungicides and herbicides such as epoxiconazole and diflufenican. Agroecological transition
544 and environmental policies encourage the protection and extension of seminatural habitats in
545 agricultural landscapes to promote biodiversity and ecosystem services. We strongly recommend that
546 the potential of these habitats to expose nontarget organisms to CUPs, the associated risk and the
547 mitigation of actual CUP contamination be considered.

548 Further, to mitigate the contamination of both off-field areas and nontarget arable soils, we advise to
549 view pesticide use reduction at landscape scale i.e., considering the surfaces and location of treated
550 crops versus other land covers within the mosaic.

551 Bernhardt et al. (2017) showed that the increases in synthetic chemicals (pesticides, pharmaceuticals,
552 and other synthetic chemicals) in terms of their total quantities, diversity, and geographic expansion
553 over the past four decades have exceeded the rate of the increase in most well-recognized drivers of
554 global change, such as rising atmospheric CO₂ concentrations, habitat destruction, and biodiversity
555 loss. Despite this situation, far less attention has been devoted to studies addressing synthetic
556 chemicals than to studies about other agents of global change, and far less funding has been dedicated
557 to this topic. This represents a critical knowledge gap with regard to scientific advances in global
558 ecology and achieving the goals of sustainable development.

559

560 **Abbreviations**

561 CUPs: currently used pesticides

562 LC50: lethal concentration 50%

563 LOD: limits of detection

564 LOQ: limits of quantification

565 NOEC: no observed effect concentration

566 PECs: predicted environmental concentrations in soils

567 RD: recommended dose

568 $TER_{\text{earthworm}}$: toxicity/exposure ratio for earthworms

569 ZA-PVS: Zone Atelier Plaine & Val de Sèvre

570

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580

581 **Author Contributions**

582 C.P. and C.F. coordinated the research project and designed the experiments. C.P., C.F., and V.B.

583 organized the sampling design. C.P. carried out the sampling. C.B. managed data curation and

584 extraction of land use metrics from maps of ZA-PVS in Geographic Information Systems. C.B. and

585 C.F. prepared the databases. C.P., C.F., and C.B. analyzed the data. C.P., C.F. and M.C. wrote the first

586 draft of the manuscript. G.D., F.L., and E.V. developed the analytical method for pesticide

587 measurements, performed the analyses and, with P.B and S.N., wrote Appendix S1. S.G. and V.B.

588 supervised the ZA PVS sampling area and provided information on land use and farming practices. All

589 authors contributed to the writing of the final version of the manuscript, which was revised for English

590 by American Journal Experts (AJE®).

591

592 **Additional information**

593 List of Supplementary materials:

- 594 - Table S1. Persistence, sorption and recommended doses of the pesticides for cereals or other
595 common crops in the studied area.
- 596 - Table S2. Mean (\pm SD) numbers of pesticides (all classes, herbicides, fungicides, and
597 insecticides) and concentrations of the five most frequent pesticides in soils and earthworms
598 according to farming management.
- 599 - Table S3. Predicted environmental concentrations in soils (PEC_s) and toxic thresholds for
600 earthworms for all pesticides.

601 Data accessibility: The full dataset will be made available as soon as the article is accepted.

602

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Table 1. Number of treated and untreated sampling site locations for soils and earthworms (*Allolobophora chlorotica*). OF means organic farming.

	Soils			Earthworms		
	Cereal crops	Grasslands	Hedgerows	Cereal crops	Grasslands	Hedgerows
Treated	53	34	0	45	30	0
Untreated	7	11 in OF 15 permanent	60	7	10 in OF 12 permanent	51
Total	60	60	60	52	52	51

Table 2. Concentrations of the 31 pesticides in the 180 soils, ordered by decreasing numbers of detections. nd for not detected. OF for organic farming. Recommended doses for cereals *or other crops* (including potential multiapplications) based on e-phy database (<https://ephy.anses.fr>). For more detail, see

Rank	Name	Type	Recommended dose (ng g ⁻¹)	Number of detected samples	Concentration max (ng g ⁻¹)	Median concentration by habitat (ng g ⁻¹)				
						Cereal crops		Grasslands		Hedgerows
						Conventionnal	OF	Conventionnal	OF	
1	Diflufenican	Herbicide	250	162	1360.7	137.3	nd	0.8	0.4	2.3
2	Imidacloprid	Insecticide	168	160	160.0	15.1	0.9	0.4	0.3	1.8
3	Boscalid	Fungicide	467	155	1211.9	4.7	2.0	0.7	0.3	3.2
4	Epoxiconazole	Fungicide	153	145	283.0	34.6	4.9	1.1	0.5	2.7
5	Prochloraz	Fungicide	600	96	485.2	0.6	nd	0.2	nd	nd
6	Napropamide	Herbicide	1680	94	19.7	0.2	0.1	nd	nd	0.1
7	Cyproconazole	Fungicide	133	82	245.8	0.3	nd	nd	nd	nd
8	Metazachlor	Herbicide	1333	75	4.2	0.2	nd	nd	nd	nd
9	S-metolachlor	Herbicide	2000	65	8.3	nd	nd	nd	nd	nd
10	Metrafenone	Fungicide	200	61	187.1	0.2	nd	nd	nd	nd
11	Pendimethalin	Herbicide	1540	57	923.1	nd	nd	nd	nd	nd
12	Pyraclostrobin	Fungicide	221	56	53.9	0.1	nd	nd	nd	nd
13	Propiconazole	Fungicide	167	47	87.1	nd	nd	nd	nd	nd
14	Aclonifen	Herbicide	1200	41	34.5	nd	nd	nd	nd	nd
15	Clomazone	Herbicide	159	39	1.0	nd	nd	nd	nd	nd
16	Thiamethoxam	Insecticide	53	37	2.0	nd	nd	nd	nd	nd
17	Pirimicarb	Insecticide	334	35	1.4	nd	nd	nd	nd	nd
18	Metconazole	Fungicide	120	28	75.2	nd	nd	nd	nd	nd
19	Thiacloprid	Insecticide	83	25	1.4	nd	nd	nd	nd	nd
20	Fluoxastrobin	Fungicide	266	25	8.6	nd	nd	nd	nd	nd
21	Dimethachlor	Herbicide	1000	16	1.5	nd	nd	nd	nd	nd
22	Pyroxulam	Herbicide	25	15	99.1	nd	nd	nd	nd	nd
23	Cloquintocet-mexyl	Herbicide safener	25	14	15.4	nd	nd	nd	nd	nd
24	Acetochlor	Herbicide	2447	12	48.8	nd	nd	nd	nd	nd
25	Cypermethrin	Insecticide	33	5	50.9	nd	nd	nd	nd	nd
26	Fenpropidin	Fungicide	1498	3	92.8	nd	nd	nd	nd	nd
27	Tau-fluvalinate	Insecticide	96	2	1.6	nd	nd	nd	nd	nd
28-31	Lambda-cyhalothrin, Bifenthrin, Deltamethrin, Cycloxydim : nd									

Table 3. Concentrations of the 31 pesticides in the 155 earthworms (*A. chlorotica*), ordered by decreasing numbers of detections. nd for not detected. <LOQ lower than the limit of quantification.

Rank	Name	Type	Number of detected samples	Concentration max (ng g ⁻¹)	Median concentration by habitat (ng g ⁻¹)				
					Cereal crops		Grasslands		Hedgerows
					Conventionnal	OF	Conventionnal	OF	
1	Imidacloprid	Insecticide	122	777.0	340	33.2	14.35	nd	35.3
2	Diflufenican	Herbicide	97	3863.0	68.6	nd	<LOQ	nd	1.9
3	Cyproconazole	Fungicide	69	117.0	nd	nd	nd	nd	nd
4	Epoxiconazole	Fungicide	64	203.0	10.3	<LOQ	nd	nd	nd
5	Thiacloprid	Insecticide	53	42.1	nd	nd	nd	nd	nd
6	Prochloraz	Fungicide	33	1210.0	nd	nd	nd	nd	nd
7	Pendimethalin	Herbicide	24	10765.0	nd	nd	nd	nd	nd
8	Boscalid	Fungicide	20	19.8	nd	nd	nd	nd	nd
9	Propiconazole	Fungicide	18	212.0	nd	nd	nd	nd	nd
10	Metrafenone	Fungicide	17	37.0	nd	nd	nd	nd	nd
11	Pyroxsulam	Herbicide	11	470.0	nd	nd	nd	nd	nd
12	Napropamide	Herbicide	5	24.0	nd	nd	nd	nd	nd
13	Fenpropidin	Fungicide	3	11.8	nd	nd	nd	nd	nd
14	Pyraclostrobin	Fungicide	3	49.7	nd	nd	nd	nd	nd
15	S-metolachlor	Herbicide	2	2.6	nd	nd	nd	nd	nd
16	Metconazole	Fungicide	2	54.6	nd	nd	nd	nd	nd
17	Fluoxastrobin	Fungicide	2	3.7	nd	nd	nd	nd	nd
18	Metazachlor	Herbicide	1	<LOQ	nd	nd	nd	nd	nd
19-31	Pirimicarb, Lambda-cyhalothrin, Cypermethrin, Thiamethoxam, Bifenthrin, Tau-fluvalinate, Deltamethrin, Clomazone, Dimethachlor, Aclonifen, Acetochlor, Cycloxydim, Cloquintocet-mexyl : nd								

Table 4. Mean (\pm SD) number of pesticides (all classes, herbicides, fungicides, and insecticides) and concentrations of the five most frequent pesticides in soils and earthworms according to the habitat (i.e., cereal fields, grasslands, or hedgerows). For cereal fields, n = 7 under organic farming (nontreated). For grasslands, n = 11 (for soils) and 10 (for earthworms) under organic farming, while n = 15 (for soils) and 12 (for earthworms) in permanent grasslands, for total numbers of 26 (for soils) and 22 (for earthworms) nontreated grasslands. Nonparametric Kruskal-Wallis tests were used for all variables, except for the total numbers in soil (ANOVA). Different letters indicate significant differences at p = 0.05 between habitats (one analysis per soil or earthworm variable i.e., number or concentrations of CUPs). For pesticide use and cropping system analyses, NS means not significant; *p < 0.05; **p < 0.01; ***p < 0.001 (Student or Wilcoxon tests).

Soil	Cereal (n=60)		Grassland (n=60)		Hedgerow (n=60)		Pesticide use (treated/untreated)	Cropping system (organic/conventional)
Number								
All classes of pesticides (31 analyzed)	10.97 (4.08)	b	7.53 (3.96)	a	7.37 (3.24)	a	***	***
Herbicide (12 analyzed)	4.37 (2.16)	b	2.85 (1.95)	a	2.62 (1.45)	a	***	**
Fungicide (10 analyzed)	4.87 (2.01)	b	3.38 (2.12)	a	3.38 (1.79)	a	***	***
Insecticide (9 analyzed)	1.73 (0.88)	b	1.30 (0.70)	a	1.37 (0.76)	a	**	**
Concentration								
Diflufenican	258.04 (346.44)	c	1.16 (1.33)	a	28.15 (90.27)	b	***	***
Imidacloprid	20.48 (22.97)	c	1.41 (2.72)	a	7.02 (22.13)	b	***	***
Boscalid	88.39 (212.31)	b	2.51 (4.39)	a	8.97 (13.47)	b	NS	NS
Epoxiconazole	51.26 (60.17)	b	6.76 (17.59)	a	9.37 (25.36)	a	***	**
Prochloraz	23.18 (83.22)	b	0.22 (0.34)	a	0.82 (2.85)	a	***	**
Earthworms								
	Cereal (n=52)		Grassland (n=52)		Hedgerow (n=51)			
Number								
All classes of pesticides (31 analyzed)	5.04 (2.25)	b	2.31 (1.74)	a	3.22 (1.65)	a	***	***
Herbicide (12 analyzed)	1.42 (1.02)	b	0.54 (0.70)	a	0.75 (0.66)	a	***	***
Fungicide (10 analyzed)	2.42 (1.56)	b	1.00 (1.03)	a	1.04 (1.08)	a	***	***
Insecticide (9 analyzed)	1.19 (0.53)	b	0.77 (0.70)	a	1.43 (0.64)	b	NS	**
Concentration								
Imidacloprid	327.55 (212.57)	b	52.76 (81.40)	a	83.81 (130.53)	a	***	***
Diflufenican	306.91 (739.79)	b	4.29 (17.63)	a	20.87 (66.93)	a	***	***
Cyproconazole	5.24 (17.53)	a	0.93 (2.29)	a	3.71 (16.33)	a	NS	*
Epoxiconazole	20.16 (36.88)	b	6.65 (26.17)	a	0.58 (2.33)	a	***	***
Thiacloprid	1.27 (6.03)	ab	0.03 (0.09)	a	1.37 (5.07)	b	NS	NS

Table 5. Environmental risk characterization based on predicted environmental concentrations in soils (PEC_{soil}) and toxicity/exposure ratio (TER) for earthworms.

	Number of pesticides for which $[C]_{soil} > PEC_{soil}$ or $TER_{earthworm} \leq$ trigger value	Number (and %) of samples for which $[C]_{soil} > PEC_{soil}$ or $TER_{earthworm} \leq$ trigger value	Pesticides of concern (number of soil samples containing each pesticide)
PEC_{soil} initial	7	40 (22%)	Boscalid (4), Cyproconazole (6), Epoxiconazole (8), Prochloraz (2), Diflufenican (17), Pyroxsulam (2), Imidacloprid (1)
PEC_{soil} accumulated/plateau	11	170 (94%)	Boscalid (2), Cyproconazole (9), Epoxiconazole (4), Prochloraz (2), Propiconazole (5), Cloquintocet-Mexyl* (14), Diflufenican (22), Pendimethalin (5), S-Metolachlore* (65), Cypermethrine* (5), Thiamethoxam* (37)
PEC_{soil} long term (time weighted average 100 days)	5	14 (8%)	Cyproconazole (6), Metrafenone (1), Prochloraz (3), Propiconazole (1), Pyroxsulam (3)
PEC_{soil} maximum	5	32 (18%)	Boscalid (4), Cyproconazole (5), Epoxiconazole (4), Diflufenican (17), Pyroxsulam (2)
$TER_{earthworm}$ acute	0	0	
$TER_{earthworm}$ chronic	4	75 (42%)	Boscalid (6), Cyproconazole (3), Epoxiconazole (52), Imidacloprid (14)

PEC_{soil} : predicted environmental concentration in soil. Details of the values and sources are provided in Sup. Mat. Table S3.

PEC_{soil} initial not available for pyraclostrobine, cloquintocet-mexyl, s-metolachlor, cypermethrin, deltamethrin, thiacloprid, and thiamethoxam.

PEC_{soil} accumulated/plateau not available for pyraclostrobine, acetochlor, dimethachlor, metazachlor. For pesticides that were not expected to accumulate in soil, PEC_{soil} accumulated was set as "0": cloquintocet-Mexyl, s-metolachlor, cypermethrin, deltamethrin, and thiamethoxam. * values of PEC_{soil} accumulated were set at 0.

PEC_{soil} long-term: value obtained from the time-weighted average at 100 days. Not available for boscalid, epoxiconazole, pyraclostrobine, cloquintocet-mexyl, diflufenican, s-metolachlor, cypermethrin, deltamethrin, imidacloprid, lambda-cyhalothrin, tau-fluvalinate, thiacloprid, and thiamethoxam.

PEC_{soil} maximum: value cited as the maximum or used in toxicity/exposure ratios in regulation and risk assessment documents. Not available for fluoxastrobine, metconazole, pyraclostrobine, clomazone, cloquintocet-mexyl, s-metolachlor, cypermethrin, deltamethrin, thiacloprid, and thiamethoxam.

$TER_{earthworm}$ acute: $LC50 / [C]_{soil}$, trigger value = 10; LC50: lethal concentration 50%.

LC50 acute earthworms available for all pesticides

$TER_{earthworm}$ chronic: $NOEC / [C]_{soil}$, trigger value = 5. NOEC: no observed effect concentration.

NOECs (reproduction) for earthworms were not available for acetochlor, cloquintocet-mexyl, cycloxydime, dimetachlor, metazachlor, deltamethrin, and pirimicarb.

Table 6. Environmental risk characterization based on the sum of risk quotients for earthworms: number of soil samples showing each risk level for the 180 plots studied, according to the type of habitat and cropping system (CF: conventional farming, OF: organic farming).

	High risk ($\sum RQ \geq 1$)		Medium risk ($0.1 \leq \sum RQ < 1$)		Low risk ($0.01 \leq \sum RQ < 0.1$)	Negligible risk ($\sum RQ \leq 0.01$)	
	<i>n</i>	% in high risk class	<i>mean</i> $\sum RQ$	<i>max</i> $\sum RQ$	<i>n</i>	<i>n</i>	<i>n</i>
Cereal fields	51	(85%)			8	0	1
CF	48	(91%)	10	38	5	0	0
OF	3	(43%)	3	6	3	0	1
Grasslands	9	(15%)			25	20	6
CF	9	(19%)	5	12	20	15	4
OF	0				5	5	2
Hedgerows	22	(37%)	4	21	26	10	2
Total	82	(46%)			59	30	9

Figure captions

Figure 1. Frequency of the number of pesticides (all classes) per soil sample and earthworm individual.

Figure 2. Concentrations (ng g^{-1} dry weight) of pesticides (herbicides in black, fungicides in purple, and insecticides in green) in a) soils ($n = 180$) and b) earthworms ($n = 155$ individuals). LOD: limit of detection, LOQ: limit of quantification.

Figure 3. Multivariate conditional inference trees for the data on pesticide concentrations in a) soils (27 pesticides) and b) earthworms (18 pesticides). Each split is represented graphically as a branch that is labeled with the classification variable; on each branch, the bar plot shows the multivariate means of pesticide concentrations (in ng g^{-1}). Above each histogram, n is the number of sites in the leaf (group). The numbers under each histogram refer to the occurrence rank of the pesticides (see Table 1 for soils and Table 2 for earthworms). C: cereal fields, G: grasslands, H: hedgerows, OM: organic matter content. The y-axis represents the pesticide concentrations in ng g^{-1} .

Figure 1.

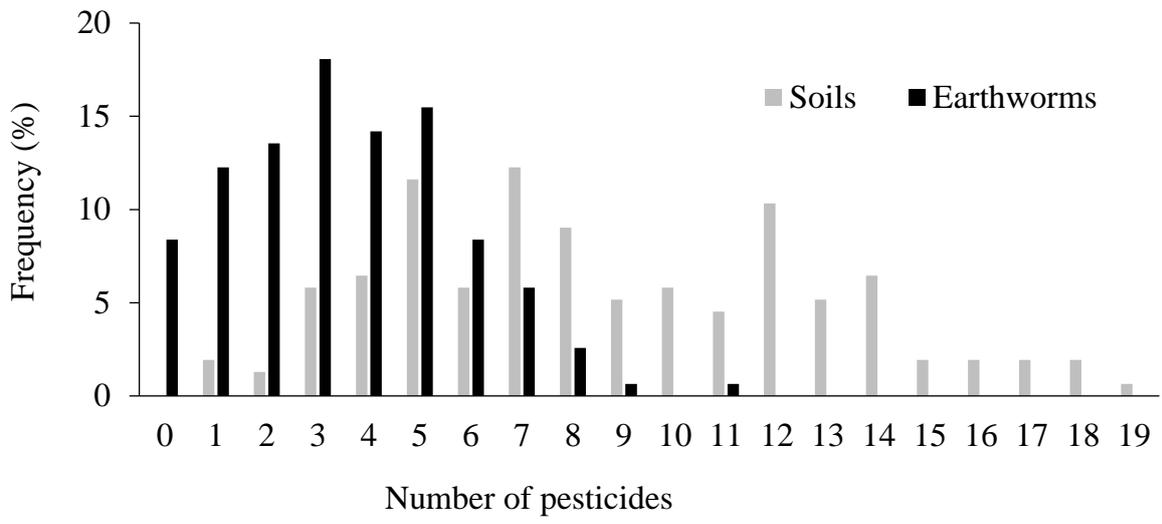
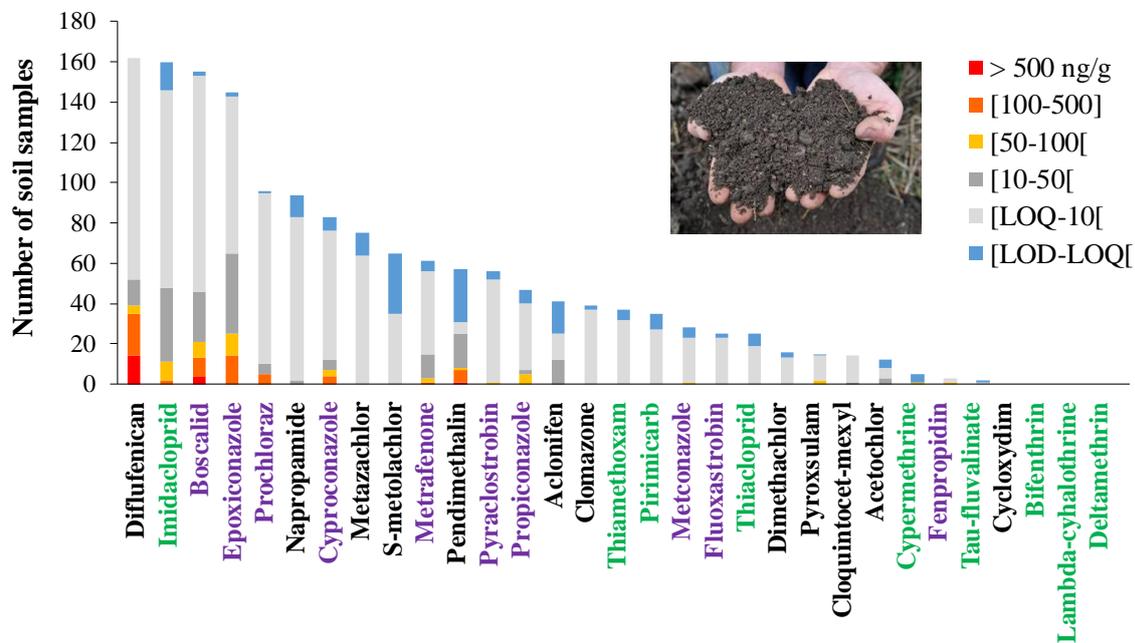


Figure 2.

a)



b)

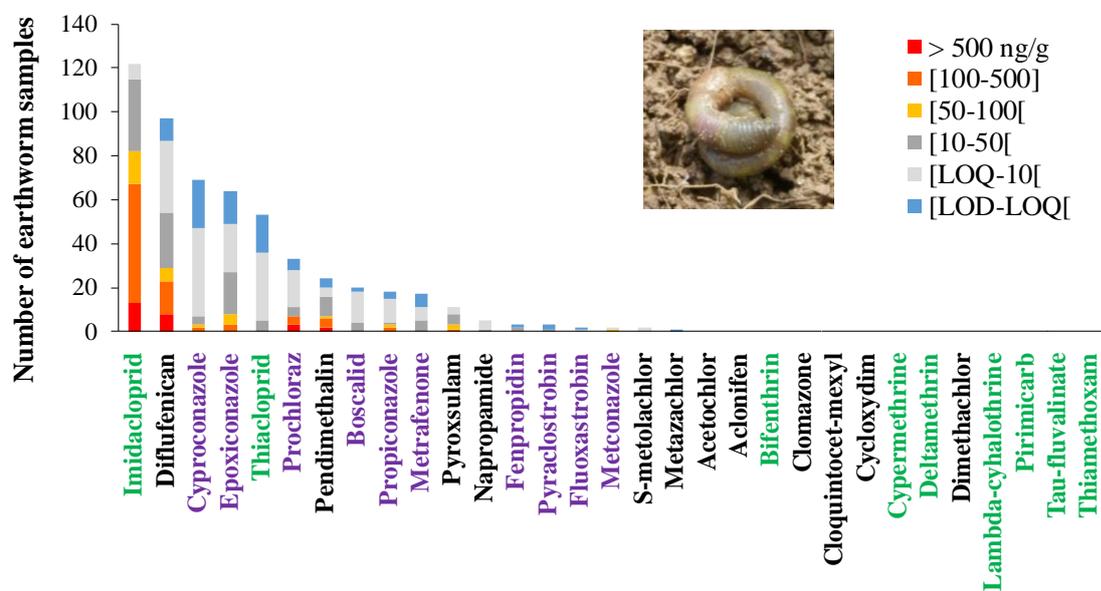
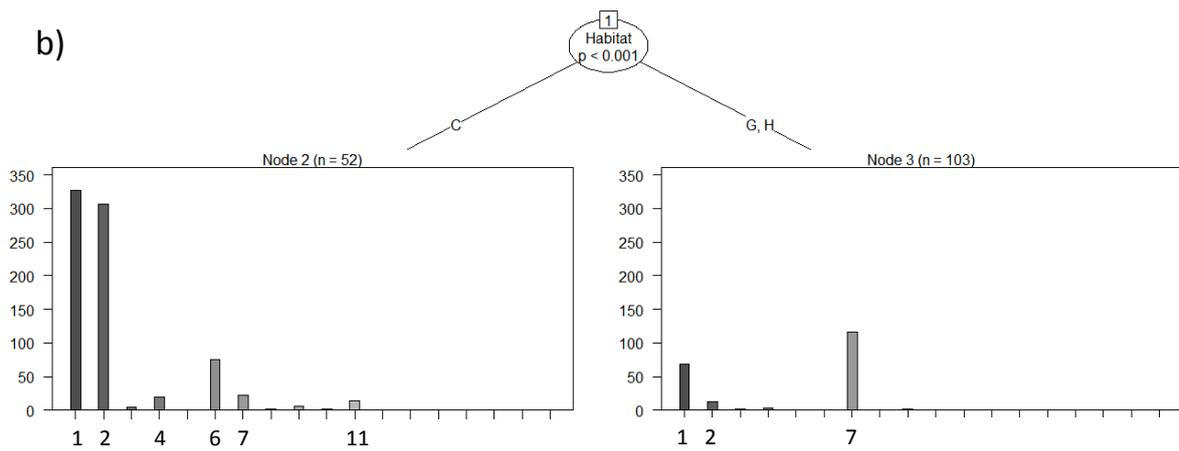
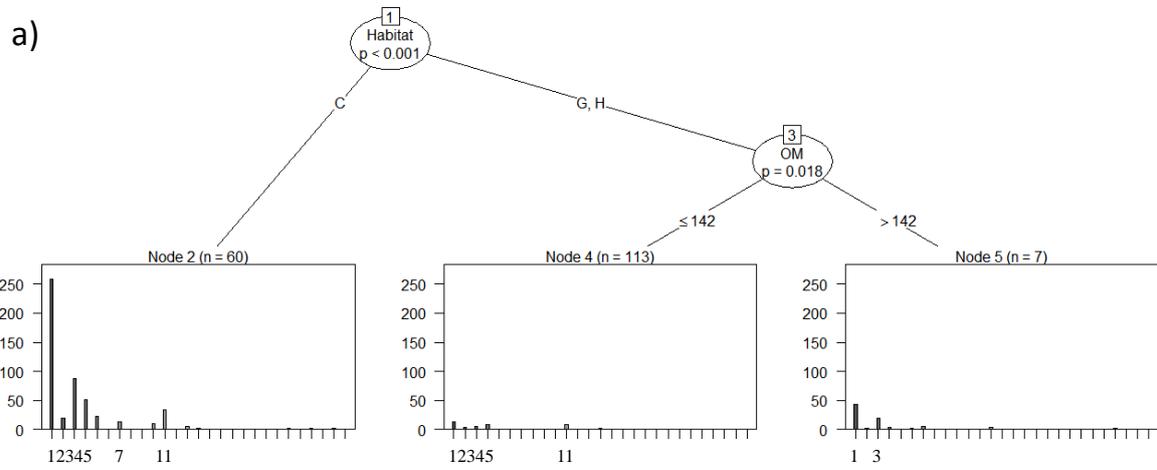


Figure 3.



Supplementary Materials

Title of the manuscript: Residues of currently used pesticides in soils and earthworms: a silent threat?

Authors : Pelosi C., Bertrand C., Daniele G., Coeurdassier M., Benoit P., Nélieu S., Lafay F., Bretagnolle V., Gaba S., Vulliet, E., Fritsch C.

Table S1. Persistence, sorption and recommended doses of the pesticides for cereals or other common crops in the studied area (x2 or x3 when several applications are recommended), according to the PPDB (DT50, K_{oc} and/or K_{foc} with 1/n) and the e-phy database (<https://ephy.anses.fr>) (recommended doses, including potential multiapplications) databases, and the limits of detection (LOD) and quantification (LOQ) in soil and earthworms. The conversion of the recommended doses from g/ha to ng/g was performed considering a 5 cm soil depth and a soil density of 1.5 g/cm³ (EFSA, 2017). *Cloquintocet-mexyl DT50: typical DT50 instead of field DT50.

Name	Type	Family	DT50 (field, days)	K_{oc} (L/kg)	K_{foc} (1/n)	Recommended dose for cereals <i>or other crops</i>		Earthworm		Soil	
						(g/ha)	(ng/g)	LOD (ng/g)	LOQ (ng/g)	LOD (ng/g)	LOQ (ng/g)
Epoxiconazole	Fungicide	Triazole	120		1073 (0.836)	115	153	0.2	0.4	0.03	0.1
Cyproconazole	Fungicide	Triazole	129		364 (0.862)	100	133	0.5	1.4	0.05	0.05
Propiconazole	Fungicide	Triazole	35.2	1086	955 (0.86)	125	167	0.3	0.7	0.01	0.01
Metconazole	Fungicide	Triazole	134.7		1116 (0.843)	90	120	0.2	0.5	0.06	0.1
Prochloraz	Fungicide	Imidazole	16.7	500	1440 (0.81)	450	600	0.2	0.4	0.01	0.03
Fenpropidin	Fungicide	- (piperidine)	49.2		3808 (0.71)	562 (x2=1124)	749 (1498)	1.2	4.1	0.1	0.4
Pyraclostrobin	Fungicide	Strobilurin	33.3	9304	9315 (0.830)	166	221	0.1	0.3	0.01	0.03
Fluoxastrobin	Fungicide	Strobilurin	52.6		848 (0.86)	100 (x2=200)	133 (266)	0.4	1.5	0.02	0.1
Metrafenone	Fungicide	Benzophenone	62	7061	3105 (0.91)	150	200	0.5	1.4	0.02	0.1
Boscalid	Fungicide	Carboxamide	254		772 (0.864)	350	467	0.5	1.6	0.01	0.1
Diflufenican	Herbicide	Carboxamide	64.6	5504	2215 (0.87)	187.5	250	0.2	0.4	0.02	0.1
Aclonifen	Herbicide	Diphenyl ether	80.4		7126 (0.922)	900	1200	1.1	3.7	0.8	2.5
Clomazone	Herbicide	Isoxazolidinone	27.3	300	128.3 (0.90)	<i>Oilseed rape: 120 g/ha</i>	159	0.1	0.2	0.02	0.02
Cycloxydim	Herbicide	Cyclohexanedione	5	59		<i>Oilseed rape, sunflower: 400 g/ha</i>	533	1.5	5.1	0.1	0.2
Acetochlor	Herbicide	Chloroacetamide	12.1	156	285 (1.21)	<i>Banned in 2013: maize 1835 g/ha</i>	2447	1	3.2	0.5	0.5
Dimethachlor	Herbicide	Chloroacetamide	3.2		69 (0.91)	<i>Oilseed rape: 750 g/ha</i>	1000	0.2	0.5	0.02	0.1
S-metolachlor	Herbicide	Chloroacetamide	23.17		200.2 (0.93)	<i>Maize: 1500 sg/ha</i>	2000	0.3	0.6	0.02	0.1
Metazachlor	Herbicide	Chloroacetamide	6.8	54	79.6 (0.993)	<i>Oilseed rape: 1000 g/ha</i>	1333	0.2	0.5	0.01	0.1
Napropamide	Herbicide	Alkanamide	72	839	885 (0.815)	<i>Oilseed rape: 1260 g/ha</i>	1680	0.2	0.5	0.02	0.1
Pendimethalin	Herbicide	Dinitroaniline	100.6	17491	13792 (0.954)	1155	1540	1.5	4.5	0.9	5.5
Pyroxsulam	Herbicide	Triazolopyrimidine	13	33.22	28.3 (0.987)	18.8	25	0.2	0.5	0.01	0.01
Cloquintocet-mexyl	Herbicide safener	Quinoleine	5*	9856		18.8	25	0.2	0.5	0.02	0.03
Imidacloprid	Insecticide	Neonicotinoid	174		225 (0.802)	126	168	0.2	0.4	0.02	0.4
Thiacloprid	Insecticide	Neonicotinoid	8.1		615 (0.875)	62.5	83	0.05	0.1	0.01	0.01
Thiamethoxam	Insecticide	Neonicotinoid	39	56.2		<i>Vine: 40 g/ha</i>	53	0.2	0.4	0.01	0.03
Cypermethrin	Insecticide	Pyrethroid	21.9	307558		25	33	9.8	32.7	0.7	0.7
Tau-fluvalinate	Insecticide	Pyrethroid	3.51	135000	186000 (0.92)	36 (x2=72)	48 (96)	1.3	4.2	0.03	0.4
Bifenthrin	Insecticide	Pyrethroid	86.8	236610		<i>Banned in 2013: cereals 10 g/ha</i>	13	0.2	0.4	0.9	0.9
Deltamethrin	Insecticide	Pyrethroid	21	10240000	10240000 (0.93)	6.25 (x2=12.5)	8 (16)	1.2	3.9	0.4	2.7
Lambda-cyhalothrin	Insecticide	Pyrethroid	26.9	283707	290311 (0.966)	7.5 (x3=22.5)	10 (30)	15.2	50.8	3.0	3.0
Pirimicarb	Insecticide	Carbamate	9		388 (0.87)	125 (x2=250)	167 (334)	0.05	0.1	0.01	0.01

Table S2. Mean (\pm SD) numbers of pesticides (all classes, herbicides, fungicides, and insecticides) and concentrations of the five most frequent pesticides in soils and earthworms according to farming management. Pesticide use includes all habitat types (grasslands, cereal fields, and hedgerows). For cropping systems, organic cereal fields and grasslands respected the rules of the AB France label.

Soil	Pesticide use		Cropping system	
	Untreated (n=93)	Treated (n=87)	Organic (n=18)	Conventional (n=102)
Total occurrence (31 analyzed)	6.78 (3.35)	10.37 (4.07)	5.56 (3.91)	9.78 (4.16)
Herbicide occurrence (12 analyzed)	2.51 (1.53)	4.10 (2.16)	2.28 (1.90)	3.84 (2.16)
Fungicide occurrence (10 analyzed)	3.05 (1.78)	4.70 (2.09)	2.33 (1.94)	4.41 (2.12)
Insecticide occurrence (9 analyzed)	1.23 (0.66)	1.56 (0.82)	0.94 (0.54)	1.53 (0.79)
Diflufenican	18.46 (73.48)	178.44 (310.78)	0.56 (0.98)	152.38 (293.63)
Imidacloprid	4.85 (18.00)	14.76 (20.96)	1.15 (2.00)	12.68 (19.99)
Boscalid	7.68 (16.76)	60.67 (180.24)	9.26 (28.93)	51.84 (167.68)
Epoxiconazole	7.11 (21.03)	38.89 (54.84)	4.74 (10.01)	33.30 (52.38)
Prochloraz	0.57 (2.31)	16.09 (70.23)	0.13 (0.30)	13.74 (65.06)
Earthworms	Untreated (n=80)	Treated (n=75)	Organic (n=17)	Conventional (n=87)
Total occurrence (31 analyzed)	2.56 (1.79)	4.55 (2.16)	1.35 (1.46)	4.13 (2.32)
Herbicide occurrence (12 analyzed)	0.59 (0.65)	1.24 (0.98)	0.18 (0.39)	1.14 (0.98)
Fungicide occurrence (10 analyzed)	0.88 (1.01)	2.15 (1.48)	0.59 (0.87)	1.93 (1.50)
Insecticide occurrence (9 analyzed)	1.10 (0.76)	1.16 (0.59)	0.59 (0.62)	1.06 (0.64)
Imidacloprid	63.79 (110.85)	252.63 (217.28)	28.04 (40.17)	221.83 (216.90)
Diflufenican	13.54 (54.15)	215.52 (625.65)	0.32 (1.16)	185.94 (585.11)
Cyproconazole	2.48 (13.10)	4.17 (14.75)	0.20 (0.59)	3.65 (13.74)
Epoxiconazole	0.45 (1.96)	18.51 (37.13)	0.05 (0.09)	16.02 (35.01)
Thiacloprid	0.88 (4.09)	0.89 (5.04)	0.03 (0.10)	0.77 (4.69)

Table S3. Predicted environmental concentrations in soils (PEC_s) and toxic thresholds for earthworms for all pesticides. LC50: lethal concentration 50%. NOEC: no observed effect concentration. PEC_{soil} accumulated/plateau: pesticides not expected to accumulate in soil, for which PEC_{soil} accumulated was set as "0". PEC_{soil} long-term: value given by the time-weighted average at 100 days. PEC_{soil} maximum: value cited as the maximum or used in toxicity/exposure ratios. NA means not available.

Class	Pesticide	PEC _{soil} initial (mg/kg)	PEC _{soil} plateau or accumulated (mg/kg)	PEC _{soil} long term (mg/kg)	PEC _{soil} maximum (mg/kg)	LC50 acute earthworm (mg/kg)	NOEC reproduction earthworm (mg/kg)	Remarks about PEC _{soil}	Sources
Fungicide	Boscalid	0.364	0.639	NA	0.364	> 500	1.197	Calculation under operating conditions defined in SANCO/11244/2011 rev. 5.	1,2
Fungicide	Cyproconazole	0.0484	0.0181	0.0450	0.0762	37.5	0.75	Calculation under scenario for cereals.	3,4
Fungicide	Epoxiconazole	0.128	0.169	NA	0.167	> 62.5	0.084	Calculation under scenario for cereals.	1,5
Fungicide	Fenpropidin	0.54	2.15	0.41	2.15	> 500	10	Calculation under scenario for cereals.	4,6
Fungicide	Fluoxastrobin	0.242	0.032	0.184	NA	> 500	1.33	Calculation under scenario for cereals.	4,7
Fungicide	Metconazole	0.094	0.15	0.083	NA	> 500	0.9	Calculation under scenario for cereals.	8
Fungicide	Metrafenone	0.196	0.843	0.179	0.843	> 500	3	Calculation under scenario for cereals.	4,9
Fungicide	Prochloraz	0.3173	0.2829	0.2068	0.6002	> 500	4.2	Calculation under scenario for cereals.	4,10
Fungicide	Propiconazole	0.0919	0.0419	0.0869	0.12	686	0.833	Calculation under scenario for Wheat and barley.	4,11
Fungicide	Pyraclostrobin	NA	NA	NA	NA	35.2	0.443	Not assumed to accumulate in soils. Indicated in soil accumulation studies and residue studies as "not required" in EU documents.	4,12
Herbicide	Acetochlor	2.8	NA	1.064	2.81	105.5	NA	Calculation under scenario for maize.	4,13
Herbicide	Aclonifen	3.2	0.3008	2.6931	3.501	150 / 97	45	Calculation under scenario for sunflower.	4,14
Herbicide	Clomazone	160	< 0.01	111.57	NA	78	> 0.8	Calculation under scenario for oilseed rape.	4,15
Herbicide safener	Cloquintocet-mexyl	NA	0	NA	NA	1000	NA	Not reported. Not assumed to accumulate in soils	4
Herbicide	Cycloxydim	0.680	0.000	0.031	0.68	> 500	NA	Calculation under scenario for beans	4,16
Herbicide	Diflufenican	0.405	0.245	NA	0.405	> 500	500	Calculation under scenario for cereals.	4,17
Herbicide	Diméthachlor	2.000	NA	0.404	2	70	NA	Calculation under scenario for oilseed rape.	4,18
Herbicide	Metazachlor	1.333	NA	0.394	1.333	219.5	NA	Calculation under scenario for oilseed rape.	4,19
Herbicide	Napropamid	2.063	0.063	1.590	3.47	282	30	Calculation under scenario for Northern Europe: Brassicas, oilseed rape for all PEC _{soil} values except for the "maximum". Value of PEC _{soil} maximum calculated for Southern Europe, under tomato cultivation	4,20
Herbicide	Pendimethalin	2.133	0.186	1.783	2.319	> 500	16,7	Calculation under scenario for cereals.	4,21
Herbicide	Pyroxulam	0.025	0.123	0.006	0.025	> 78	1.07	Calculation under scenario for cereals.	4,22

Herbicide	S-metolachlor	NA	0	NA	NA	570	< 2.54	Not assumed to accumulate in soils. Indicated in soil accumulation studies and residue studies as “not required” in EU documents.	4,23
Insecticide	Bifenthrin	0.0195	0.027	0.0165	0.027	>8	1.065	Calculation under scenario for cereals.	4,24
Insecticide	Cypermethrin	NA	0	NA	NA	> 100	> 5.3	No accumulation observed in field studies, no accumulation/residue in soil studies required as cited in SANCO documents	4,25
Insecticide	Deltamethrin	NA	0	NA	NA	> 1290	NA	Not assumed to accumulate in soils. Indicated in soil accumulation studies and residue studies as “not required” in EU documents.	4,26
Insecticide	Imidacloprid	0.156	0.184	NA	0.184	10.7	> 0.178	Calculation under scenario for sugar beet.	4,27
Insecticide	Lambda-Cyhalothrin	0.0145	0.0157	NA	0.0431	> 500	3.125	Calculation under scenario for wheat.	4,28
Insecticide	Pirimicarb	0.276	0.198	0.221	0.338	> 60	NA	Calculation under scenario for wheat.	4,29
Insecticide	Tau-fluvalinate	0.03596	NA	NA	0.09	110	1.44	Calculation under scenario for wheat.	4,30
Insecticide	Thiacloprid	NA	0.14	NA	NA	105	< 62.5	Not assumed to accumulate in soils. Indicated in soil accumulation studies and residue studies as “not required” in EU documents.	4,31
Insecticide	Thiamethoxam	NA	0	NA	NA	> 1000	5.34	Not assumed to accumulate in soils. Indicated in soil accumulation studies and residue studies as “not required” in EU documents.	4,32

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