



# *A review of the factors that influence pesticide residues in pollen and nectar: future research requirements for optimising the estimation of pollinator exposure*

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1 **A review of the factors that influence pesticide residues in pollen and nectar:**  
2 **future research requirements for optimising the estimation of pollinator**  
3 **exposure**

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10 **Abstract**

11 In recent years, the impact of Plant Protection Products (PPPs) on insect pollinator decline has  
12 stimulated significant amounts of research, as well as political and public interest. PPP residues have  
13 been found in various bee-related matrices, resulting in governmental bodies worldwide releasing  
14 guidance documents on methods for the assessment of the overall risk of PPPs to different bee  
15 species. An essential part of these risk assessments are PPP residues found in pollen and nectar, as  
16 they represent a key route of exposure. However, PPP residue values in these matrices exhibit large  
17 variations and are not available for many PPPs and crop species combinations, which results in  
18 inaccurate estimations and uncertainties in risk evaluation. Additionally, residue studies on pollen and  
19 nectar are expensive and practically challenging. An extrapolation between different cropping  
20 scenarios and PPPs is not yet justified, as the behaviour of PPPs in pollen and nectar is poorly  
21 understood. Therefore, this review aims to contribute to a better knowledge and understanding of  
22 the fate of PPP residues in pollen and nectar and to outline knowledge gaps and future research needs.  
23 The literature suggests that four primary factors, the crop type, the application method, the  
24 physicochemical properties of a compound and the environmental conditions have the greatest

25 influence on PPP residues in pollen and nectar. However, these factors consist of many sub-factors  
26 and initial effects may be disguised by different sampling methodologies, impeding their exact  
27 characterisation. Moreover, knowledge about these factors is ambiguous and restricted to a few  
28 compounds and plant species. We propose that future research should concentrate on identifying  
29 relationships and common features amongst various PPP applications and crops, as well as an overall  
30 quantification of the described parameters; in order to enable a reliable estimation of PPP residues in  
31 pollen, nectar and other bee matrices.

## 32 **Keywords**

33 Pesticides; Risk Assessments; Pollinator; Pollen and Nectar; Residues

## 34 **Capsule**

35 Pesticide residue values within pollen and nectar have potentially significant consequences for the  
36 reliability of risk assessments for wild and managed bee populations, however, the reasons and  
37 mechanisms underlying variations in residues are poorly understood and require greater  
38 investigation.

## 39 **Introduction**

### 40 **Usage, benefits and drawbacks of Plant Protection Products**

41 The global population has increased rapidly, tripling since 1950 to a current total of 7.6 billion  
42 (Population Reference Bureau 2017), and is predicted to expand to 9.6 billion by 2050 (UN 2017). This  
43 growth has been facilitated by the intensification of crop production as a result of new developments  
44 and innovations (Carvalho 2006; Johnson 2000). As a consequence, the daily food supply per capita  
45 increased from 2196 kcal day<sup>-1</sup> in 1960 to 2884 kcal day<sup>-1</sup> in 2013, with cereal yields almost tripling in  
46 the same time period (FaoStat 2017). Concurrent increases in production, use and trade of Plant  
47 Protection Products (PPPs) indicate their contribution to these increases in food production (Atwood  
48 and Paisley-Jones 2017; Gilland 2002; Tilman 1999; Tilman et al. 2001; Zhang et al. 2011). Today,

49 approximately 1000 active ingredients (a.i.s) (i.e. the components in PPPs which are active against  
50 pests/plant diseases) are globally available (Lewis et al. 2016).

51 The predominant use of PPPs is in the agricultural sector to protect crops from weeds, fungal  
52 pathogens and pests (Wilson and Tisdell 2001). Estimates suggest the losses in plant production  
53 without PPPs would be up to 80% for some crops with potentially severe economic consequences  
54 (Oerke and Dehne 2004; Oliveira et al. 2014; Pimentel 1997). Outside of the agricultural sector, PPPs  
55 are a cost and labour efficient method for the protection and maintenance of public spaces, for  
56 example weed control on railways and streets (Cooper and Dobson 2007). In the future, the targeted  
57 use of PPPs could further grow in importance; consequences of globalisation and climate change are  
58 predicted to change the distribution and life cycles of many pest species, which could render previous  
59 control strategies ineffective (Hulme 2017; Rosenzweig et al. 2001). Therefore, there is a strong  
60 argument to suggest that PPPs currently make a significant contribution to stable and reliable crop  
61 yields, high food quality and the prevention of economic losses, which is a key factor in enabling the  
62 global food system to continue to operate in its current format.

63 Nevertheless, PPPs are toxic chemicals and, in the absence of mitigation, some exposure to non-target  
64 organisms and the ecosystem is inevitable. Due to their wide range of applications, PPP residues and  
65 their metabolites can be found in many ecosystems, with the potential to cause various effects on  
66 humans, soil and water organisms, birds, mammals and invertebrates (Mostafalou and Abdollahi  
67 2017; Pimentel 2005; Tilman 1999).

### 68 **PPPs and insect pollinators**

69 In recent years, high overwintering losses of honey bee colonies and declines in populations of other  
70 insect pollinator species in Europe and North America (Lee et al. 2015; Ollerton 2017; Potts et al.  
71 2010b; Seitz et al. 2016) have raised concerns about the contribution of PPPs to these losses (IPBES  
72 2016). Managed and wild pollinator species provide vital ecosystem services, particularly for agro-  
73 ecosystems (Albrecht et al. 2012; Klein et al. 2007; Vanbergen et al. 2014; Veddeler et al. 2008) and

74 Gallai et al. (2009) calculated the total economic value of pollination worldwide to be € 153 billion. As  
75 a result, the toxic effects of PPPs on pollinators, particularly neonicotinoids, has become the focus of  
76 significant amounts of research, and political and public interest. There is a broad consensus amongst  
77 researchers in the field that declines are the result of a combination of factors including habitat loss,  
78 pests/diseases and PPPs (Goulson et al. 2015; IPBES 2016; Potts et al. 2010a). Whilst the overall role  
79 of PPPs on pollinator declines is still debated, there is clear evidence for both the exposure of bees to  
80 a range of chemical products via contact and oral exposure (e.g. Botias et al. 2017; Chauzat et al. 2010;  
81 Johnson et al. 2010; Kiljanek et al. 2017; Tosi et al. 2018) and the toxicity of PPPs to bees in laboratory  
82 toxicity studies (e.g. Kasiotis et al. 2014; Pettis et al. 2012; Sanchez-Bayo et al. 2017; Woodcock et al.  
83 2017; Wu et al. 2011).

84 Overall, there is a difficult trade-off between permitting the use of products upon which modern  
85 agriculture relies for the protection of crops and maintaining vital environmental goods and services,  
86 which themselves have an important role in sustainable food production. Therefore, in order to ensure  
87 the safety of PPPs, complex and highly regulated processes of environmental risk assessments have  
88 been developed.

89 With respect to pollinators, the European Food Safety Authority (EFSA 2013) published a Guidance  
90 Document on the risk assessment of PPPs on bees (including honey bees, *Apis mellifera* L.,  
91 Bumblebees, *Bombus spp.* and solitary bee species), to outline a process by which PPPs can be  
92 evaluated for their potential risks in causing unacceptable harm to bees. Similar approaches have been  
93 published in the US, Canada and Brazil (Cham et al. 2017; USEPA 2014). An important component in  
94 these approaches are PPP residue levels in pollen and nectar. They represent a key route of exposure  
95 for pollinators as, in many species, all life stages feed to some extent upon these food sources (Rortais  
96 et al. 2017; Villa et al. 2000). However, knowledge to enable a more accurate prediction of PPP  
97 residues in pollen and nectar is limited and a number of barriers, which are discussed in more detail  
98 in the next section, inhibit a clear assessment of residue levels used in risk estimation.

## 99 **Aim of the review**

100 In this review our aim was to identify and compile existing literature data on the behaviour and fate  
101 of residues in pollen and nectar following PPP applications, and outline the manifold parameters which  
102 appear to influence these residues. In doing so we identify knowledge gaps concerning the variability  
103 of PPP residue values in pollen or nectar and highlight future research needs, in order to enable a  
104 precise prediction of residue levels for pollinator risk assessments in future and to encourage, initiate  
105 and facilitate further research in this field.

## 106 **Pollinator risk assessments and evidence base**

### 107 **Current methodological approaches for pollinator risk assessments**

108 Current approaches for pollinator risk assessments (e.g. Cham et al. 2017; EFSA 2013; USEPA 2014)  
109 pursue similar strategies and methodologies. In general, effect studies (e.g. laboratory adult acute oral  
110 toxicity studies, larvae toxicity studies) and exposure estimates (contact or oral) are combined in a  
111 tiered approach to assess the risk of PPPs to pollinators, ranging from very conservative estimates to  
112 more realistic scenarios. While the latter requires high data input and more extensive studies, in the  
113 lower, more conservative tiers, worst-case default values can be applied. Theoretically, such an  
114 approach allows for more rapid and cost-effective initial assessments that are robust enough to  
115 separate those PPPs that pose a potential risk to bees from those that can be considered of low risk.  
116 To assess the risk from oral exposure of bees to PPPs, the guidance documents (Cham et al. 2017;  
117 EFSA 2013; USEPA 2014) provide general default residue values in pollen and nectar for different  
118 application scenarios, which aim to be protective. If the assessment fails in lower tiers and risk  
119 mitigation is not possible, the guidance documents cited above suggest a refinement of the  
120 assessment in higher tiers, for example by using representative “real” PPP residue values in pollen or  
121 nectar or compound and crop specific data, which can be further refined by conducting field trials.

### 122 **Barriers associated with PPP residues in bee products and their implementation in risk assessments**

123 A recent proposal made in reference to EFSA's risk assessment from the European Crop Protection  
124 Authority (ECPA 2017), which represents the industrial sector, concluded that the current guidance is  
125 over-conservative and that even substances known to be non-toxic to bees fail at lower tiers. They  
126 assert that, in most cases, a higher tier refinement is required. In order to facilitate higher tier  
127 assessments, oral exposure estimates must be refined using representative residue data (Cham et al.  
128 2017; EFSA 2013; USEPA 2014). However, data on residue levels in floral resources vary widely and  
129 are unknown for many PPPs and crop species (EFSA 2013; Lundin et al. 2015). Table 1 provides a brief  
130 overview of PPP residue data recorded in pollen from spray applications, illustrating the variation of  
131 PPP residues from different active ingredients and in different crops. These data are taken from a  
132 recent meta-study (Kyriakopoulou et al. 2017) and from the pollinator risk assessment published by  
133 EFSA (2013), both providing a comprehensive overview and summary of data on the available residue  
134 data in bee-relevant matrices and products, which were gathered from Draft Assessment Reports  
135 (DARs), literature and peer reviews of active ingredients.

136 Overall, there is wide variation in residues, with differences not only between various active  
137 ingredients and crop combinations, but also within each of these groups. For instance, aggregated  
138 residues from various PPPs vary considerably not only within *Brassica* pollen, but also from uses of  
139 individual PPPs, such as teflubenzuron on *Brassica*. Similar findings can be observed for residues in  
140 nectar (Table S1).

141 Both publications highlighted the fact that the available studies differed considerably in design,  
142 sampling timing, sampling methodology and application scenarios, or lacked data for certain types of  
143 active ingredients. Thus residue data is difficult to compare. Overall, knowledge about PPP residues in  
144 pollen or nectar is fragmentary and only a small proportion of treatments and crops have been taken  
145 into account, with the majority of residue values provided for neonicotinoids and oilseed rape (OSR)  
146 (*Brassica napus* L.). Pollinator risk evaluation is therefore based on extrapolated residue data and as a  
147 consequence, on an incomplete dataset. However, the knowledge regarding PPP residues present in



148 pollen and nectar is too limited to allow extrapolations or conclusions to be drawn from those crops  
149 where data are available.

150 Yet, if risk assessments are based on residue values that are not representative for the treatment  
151 scenario and cropping system, the risk posed from PPPs to pollinators might be incorrectly estimated  
152 (Lundin et al. 2015), resulting in false negatives (i.e. misuses of concern), or in false positives, which  
153 may result in unnecessary higher tier testing. The currently available data sets can neither mitigate  
154 the variability and incompleteness, nor rationalise how this should be addressed in risk assessments  
155 or why these variations in PPP residues occur.

#### 156 **PPP residue studies in pollen and nectar**

157 Extensive studies are needed to derive reliable PPP residue values in pollen and nectar. However, the  
158 exact determination of residues in bee-attractive plants is expensive and time consuming. Relatively  
159 large volumes of the target matrices are required for the chemical analyses needed to quantify the  
160 PPP residues, but pollen and nectar are typically produced only in small quantities. Furthermore,  
161 numerous active substances, crop species and application scenarios must be considered. The ECPA  
162 (2017) claimed that, in order to meet the requirements of the EFSA guidance document (EFSA 2013),  
163 for a single product used on five different bee-attractive crops, up to 75 residue studies would need  
164 to be conducted, with associated costs of approximately € 7.5m. Consequently, the development and  
165 registration of new products and innovations, in addition to the re-authorisation of already approved  
166 PPPs, are likely to incur large costs, which may limit the availability of PPPs. According to the ECPA,  
167 minor use crops are most likely to be affected, which are often economically important for their  
168 growers and for crop diversity, but not of significant importance to the industry to justify high costs  
169 for research and development.

170 Furthermore, with new insights and findings becoming apparent and a better comprehension of risks  
171 posed by PPPs in recent decades, it is likely that regulatory requirements will be further increased and  
172 adapted and that applicants for active ingredient and PPP registrations, PPP producers and responsible

173 authorities will need to deliver more detailed data regarding the fate of PPPs in plant matrices  
174 important to pollinators.

175 Thus, to ensure the accurate protection of pollinators and to permit scope for developments in crop  
176 protection, methods need to be devised to enable an accurate estimation of PPP residues in pollen  
177 and other bee-important matrices that require reduced effort and expenditure. In order to achieve  
178 this, a better knowledge and understanding of the fate of PPP residues in pollen and nectar is  
179 necessary. The identification of patterns and relationships of PPP residues within the plant and  
180 between different species may provide an opportunity to identify better methods for accurate  
181 estimation of residue levels for diverse PPPs and cultivation methods. However, little is understood  
182 about the behaviour and relationship of residues in floral resources, which can be altered and  
183 influenced by numerous factors.

184

#### 185 **Factors influencing PPP residues in pollen, nectar and other related matrices**

186 An assessment of the literature suggests that there are four primary factors which could influence the  
187 level of PPP residues in pollen and nectar and other related matrices: i) crop related parameters ii)  
188 discrepancies in PPP application method, timing and dose rate iii) physicochemical properties of the  
189 active ingredient and iv) environmental conditions. These primary factors consist in turn of several  
190 sub-factors which can all potentially contribute to variable PPP residues in pollen or related crop  
191 matrices. The first two factors listed are considered more often in the literature, since they are  
192 tangible and relatively easy to determine under constant conditions. By contrast, the effects of  
193 environmental conditions are more difficult to isolate, as they can, for example, influence the chemical  
194 properties of an active ingredient, as well as the development and physiology of a plant. Hence, there  
195 are a wide range of factors influencing PPP residue levels in pollinator relevant matrices, which are  
196 strongly interdependent and form a complex system. Another factor which can unintentionally  
197 influence the results of PPP residue levels in pollen and nectar is the sampling methodology. For

198 instance, OSR flowers are often collected and then incubated for a certain time period and  
199 temperature to facilitate pollen release (e.g. Botias et al. 2015). The loss of water might result in higher  
200 PPP concentrations, but conversely the high temperatures can initiate a dissipation of PPP residues.  
201 In other studies pollen is collected by grinding the anthers to powder (e.g. Jiang et al. 2018), collecting  
202 pollen in boxes as it falls naturally from plants (Schmuck et al. 2001) or by using bees (e.g. Choudhary  
203 and Sharma 2008). These discrepancies in sampling are often not scrutinised in studies but might  
204 influence the comparability of results.

### 205 **General findings**

206 Although high variability is typically observed in PPP residues in pollen and nectar, there are some  
207 instances that permit comparisons among a wide range of PPPs/crop systems. Kyriakopoulou et al.  
208 (2017) detected statistically significant differences among sampling matrices, with the residue levels  
209 in both pollen and nectar being highest when extracted directly from flowers than from bees. Such  
210 differences could be caused by dilution effects, when bees mix pollen from untreated and treated  
211 crops (Bonmatin et al. 2015; Rolke et al. 2016). In many studies a dilution effect, cross contamination  
212 from other fields (e.g. Kunz et al. 2015) or chemical alterations cannot be excluded when pollen and  
213 nectar is collected by free flying bees and it is often difficult to directly link the residues found to the  
214 previous PPP treatment, unless studies are conducted using bee-sampled pollen collected from  
215 tunnelled crops (i.e. no alternative sources of pollen are available).

216 Furthermore, Kyriakopoulou et al. (2017) detected higher residues in pollen than in nectar, a  
217 phenomenon which has been reported in several other studies, which employ a range of treatment  
218 regimens (e.g. Choudhary and Sharma 2008; Cowles and Eitzer 2017; Dively and Kamel 2012; EFSA  
219 2012; Goulson 2013; Jiang et al. 2018). Reasons for this difference have not been investigated thus  
220 far; however, several possible mechanisms can be proposed. If bee- collected matrices are analysed,  
221 the effect could be caused by the partial metabolism of residues in nectar within the bees (Gong and  
222 Diao 2017; Sanchez-Bayo and Goka 2014). However, similar results have also been reported from

223 samples taken directly from the plant. Cowles and Eitzer (2017) suggested a relationship between  
224 residue levels in pollen and nectar and whether nectaries and anthers are supplied by phloem or  
225 xylem. Choudhary and Sharma (2008) presumed analytical impediments, for example the active  
226 ingredient could form conjugates with sugars in nectar, thus becoming difficult to extract, or that, due  
227 to morphological differences, there may be differences in either the initial levels of PPPs or in their  
228 rates of degradation. Overall, the meta-analysis by Kyriakopoulou et al. (2017) found a strong  
229 correlation between the residue levels in pollen and nectar, though none of the individual studies  
230 included in the meta- analysis has directly compared this parameter thus far.

### 231 **Crop related parameters**

232 Although few crop species are considered in studies on PPP residues in pollen and nectar, there is  
233 evidence that crop traits have an influence on the residue levels in bee-important matrices.  
234 Differences in residue levels in various plant parts can be explained by a dilution effect with plant  
235 growth (more biomass) (Holland et al. 1996), plant height (Kleier 1994) and even plant age (Bonmatin  
236 et al. 2015), for example when PPPs have the ability to be adsorbed to plant compounds like lignin  
237 (Fujisawa et al. 2002). Overall, these effects are strongly related to the physicochemical properties of  
238 a compound (see section below for full discussion on the effects of physicochemical properties). Soil  
239 treatments of the systemic compound imidacloprid demonstrated that there is a clear gradient with  
240 respect to residue levels from the leaves at the bottom of the plant up to the leaves at the top of the  
241 plant, and eventually to the flowers and pollen (Alsayeda et al. 2007; Bonmatin et al. 2005; Johnson  
242 2012; Laurent and Rathahao 2003; Stoner and Eitzer 2012).

243 This raises questions as to whether conclusions drawn from the PPP residue levels found in foliage can  
244 be applied to those in pollen/nectar and whether crops with lower biomass exhibit higher residue  
245 levels in leaves and consequently in pollen or nectar. Balfour et al. (2016) found that neonicotinoid  
246 concentrations in the tissues of flowering maize (*Zea mays* L.) and OSR are negatively correlated with

247 plant mass, however, they did not directly compare these results with pollen and nectar collected  
248 from the same plots.

249 Dively and Kamel (2012) found a strong correlation of imidacloprid residues in squash (*Cucurbita pepo*  
250 L.) between leaves and pollen, and leaves and nectar ( $r = 0.94$  and  $r = 0.88$ , respectively;  $p < 0.001$ )  
251 following different soil application treatments. This, however, was analysed only during the course of  
252 one year and the trend was not replicated for metabolites of imidacloprid or other investigated  
253 neonicotinoids. Dively and Kamel (2012) suggested that the diverse chemical properties of the  
254 investigated compounds, mainly the solubility, were the reason for a varying uptake and translocation  
255 rate, and consequently higher residue levels of other neonicotinoids. However, the differences in  
256 residue levels could also be due to the fact that they randomly selected leaves for analysis during their  
257 study. Considering the dilution effect and gradient, different results might have been found by using  
258 leaves of similar size and position on the plant. Such an approach was employed by Jiang et al. (2018),  
259 who collected only newly expanded leaves of cotton (*Gossypium* sp.) over a one-month period.  
260 Although no correlations were observed in nectar, correlations between imidacloprid and  
261 thiamethoxam residues in leaves and pollen ( $r = 0.88$  and  $r = 0.90$ , respectively;  $P < 0.001$ ) were found.  
262 However, it is unclear whether these observations also apply for other crops, other PPPs (i.e. those  
263 with non-systemic properties) and different application scenarios.

264 Demonstrating similarities between species has proven to be problematic, with even varieties of the  
265 same species resulting in different residue levels. This was demonstrated by Bonmatin et al. (2003),  
266 who investigated several sunflower (*Helianthus annuus* L.) varieties after a seed treatment with  
267 imidacloprid. The final concentration in flowers was dependent on the variety, with ranges from 2.7  
268  $\mu\text{g g}^{-1}$  up to 7  $\mu\text{g g}^{-1}$ . The authors did not provide any information about habitus or other species-  
269 specific characteristics, but an acropetal decrease of residues in foliage, as described above, was  
270 detected for all varieties. In addition, during the formation of the capitula of the sunflower there was  
271 a sudden increase in imidacloprid residue levels in the upper parts of the plants. Similar findings were

272 reported by Laurent and Rathahao (2003), analysing different parts of sunflowers. Moreover, PPP  
273 concentrations in pollen were similar to those found in the floret dish. It can therefore be concluded  
274 that the pollen was contaminated by the late shift of PPP residues in sunflowers. The authors  
275 presumed a remobilisation process, in which compounds accumulated in older leaves were  
276 transferred towards the upper part of plants during the reproductive stage. However, imidacloprid is  
277 a xylem-mobile PPP; hence, it should not re-translocate (Sur and Stork 2003).

278 Laurent and Rathahao (2003) provided another explanation for the phenomenon, suggesting that it  
279 was a consequence of the differential root system of sunflowers. This consists of fascicular roots,  
280 which grow horizontally in the superficial layer of the soil, and a deeper root system; thus, various soil  
281 levels can be penetrated. Sunflowers are particularly capable of recovering PPP residues from soils,  
282 which can be attributed to this extensive root system (Bonmatin et al. 2003; Mitton et al. 2016).  
283 Consequently, the more pronounced root system of an older plant can take up more PPPs from the  
284 soil, leading to an increase of residues during the flowering period. The root system is also an  
285 important parameter concerning the PPP uptake from soil in other species, for example from the  
286 *Cucurbita* family (Otani et al. 2007). For instance, cucumbers (*Cucumis sativus*) grafted with high  
287 uptake root stocks could recover up to 70% more dieldrin (an organochlorine insecticide) than those  
288 with a low uptake root stock (Otani and Seike 2007), giving a further explanation as to why different  
289 varieties exhibit different residue levels from soil treatments. Regarding the ability of different root  
290 systems to shift the PPP residues in plants, plant density and whether experiments are conducted in  
291 field or pots might also be important parameters to understand the variability of residues in flowers  
292 and should be considered in soil-applied PPP residue studies.

293 Obviously, these observations are less relevant for foliar-sprayed or non-systemic PPPs (see section  
294 below for full discussion of the effects of application method). The PPP's chemical properties, the  
295 morphology and the structure of the leaves, flowers and cuticle determine the uptake rate of the  
296 product and hence the likelihood of translocation to pollen or nectar (DiTomaso 1999; Kirkwood 1999;

297 Price and Anderson 1985). For example, a hairy or waxy leaf structure affects the retention time of  
298 chemicals on the surface (Yu et al. 2009); this can alter the uptake rate of the PPPs and hence the  
299 chemicals' exposure to environmental conditions. Therefore, even under similar conditions, different  
300 plant species will show different residue levels and behaviour. Kyriakopoulou et al. (2017) found  
301 species-related differences in pollen and nectar residues. In particular, OSR showed the highest  
302 residue values in comparison to other plants. However, there were more data available for OSR and  
303 the majority of other species was summarised to one group. Therefore, there is limited confidence as  
304 to whether OSR genuinely is a crop which accumulates a high level of PPP residues in pollen or nectar.  
305 For a summary of this section and problems regarding pollinator risk assessments see Figure 1.

306

### 307 **Application Method, Application Timing, Dose Rate**

308 The application method, timing of the application and the dose rate of an applied PPP are strongly  
309 interdependent. For example, by using a seed treatment, the longest possible time period between  
310 application and flowering of the plant is attained. In contrast, many fungicides are sprayed directly  
311 onto the plant shortly before or during flowering, especially when they have been assessed as non-  
312 harmful for bees. Furthermore, seed dressings often contain less active ingredient per hectare and  
313 therefore may be considered to be more environmentally friendly. This is reflected in the residue  
314 levels of foliar applications and seed dressings reviewed by EFSA (2012, 2013), with residues from seed  
315 dressings being substantially lower than from spray applications. Evidence regarding the effect of dose  
316 rate on PPP residues in pollen was provided by Bonmatin et al. (2005), who used three different doses  
317 of the systemic active ingredient imidacloprid, applied as a seed dressing to sunflower seed. The  
318 concentration of imidacloprid in the capitula of several varieties became higher when the dose rate  
319 was increased. Furthermore, the ascent of imidacloprid during flowering (see section above) was more  
320 pronounced when the doses of the seed dressing were high. However, studies directly comparing the  
321 effect on residues in pollen and nectar at different dose rates of foliar applied or non-systemic PPPs

322 are scarce, although it is possible to discern a certain trend from the detailed values provided by EFSA  
323 (2012), indicating that higher dose rates cause higher residues in pollen and nectar.

324 Yet, it cannot be concluded that a high application of PPPs naturally results in a high exposure for bees  
325 or in high residues in relation to the dose rate. Choudhary and Sharma (2008) applied a range of PPPs  
326 to mustard (*Brassica juncea* Czern.) using foliar application, each with a defined dose rate, and showed  
327 that PPPs applied at higher rates indeed tend to result in higher residues in pollen and nectar (Table  
328 2). Interestingly, RUDs - the residue unit employed in risk assessments to account for different dose  
329 rates (RUD = concentration in nectar/ pollen ( $\text{mg kg}^{-1}$ ) at an application rate of  $1 \text{ kg ha}^{-1}$  or  $1 \text{ mg seed}^{-1}$ ) - exhibited an opposing trend in this experiment. Lambda-cyhalothrin afforded the highest PPP  
330 residues relative to the dose rate and endosulfan, though applied at the highest dose, afforded the  
331 lowest residues with respect to the dose. Thus, the ratio of residues from different PPPs relative to  
332 the dose rate is not equal for all compounds, it is rather influenced by other factors, for example the  
333 different chemical properties of the active ingredients, which are responsible for varying uptake and  
334 accumulation in floral parts.  
335

336 Nevertheless, Byrne et al. (2014) observed higher residues in nectar with a doubled dose rate  
337 compared to the normal dose rate when treating citrus trees with a soil drench application of  
338 imidacloprid. This effect was reinforced at later sampling dates, i.e. with a longer time period between  
339 application and flowering. It is assumed that a longer time period between application and flowering  
340 results in lower residues because of the dilution, metabolism and dissipation in plants. For  
341 imidacloprid, however, to a certain extent the contrary was shown. Whether this effect is similar to  
342 that described by Bonmatin et al. (2003) and Laurent and Rathahao (2003) in the above section is not  
343 verifiable. It does, however, illustrate that the timing of the application can have a significant impact  
344 on residue levels in pollen and nectar. These findings can also be important when comparing varieties  
345 and cropping systems. For instance, Pohorecka et al. (2012) found substantially lower residues of  
346 thiamethoxam in bee foraging products from winter OSR than spring OSR. It is hypothesised that the



347 longer time period between treatment and bloom of winter OSR led to a higher degradation of the  
348 active ingredient.

349 Cowles and Eitzer (2017) also detected late imidacloprid accumulation in sunflower pollen, but under  
350 different experimental conditions. Their extensive experimental setup considered three  
351 neonicotinoids, applied at different times with different application methods to sunflower and swamp  
352 milkweed (*Asclepias incarnata* L.). Again, a low rate imidacloprid soil drench application was the only  
353 scenario (application rate, method, and insecticide) found to result in increasing concentrations as the  
354 time post-application increased; which meant a soil drench application performed 10 weeks prior to  
355 bloom was the only timing for this application scenario that exceeded the designated “toxicity  
356 threshold” for bees in pollen concentrations at the lowest dose rate. In contrast, dinotefuran soil  
357 drench applications led to higher residues when they were applied closer to the blooming period. The  
358 authors concluded that dinotefuran has a better solubility and higher mobility than imidacloprid and  
359 therefore the uptake is faster, whereas the uptake of imidacloprid takes longer and so residues  
360 accumulate later in pollen. This finding might be especially important for the estimation of residues in  
361 crops with a pronounced short or long life cycle and shows that the physicochemical properties of a  
362 compound must always be taken into account (see section below for full discussion of physicochemical  
363 properties).

364 Cowles and Eitzer (2017) demonstrated that higher application rates resulted in higher residues in  
365 pollen and nectar, depending on the chemical applied. However, the method of application had the  
366 strongest influence on pollen and nectar residue levels. Soil drench applications resulted overall in  
367 higher residues than the foliar applications, even if both were applied only two weeks before bloom.  
368 This indicates that, even though the uptake via leaves is good, it cannot be compared with the uptake  
369 and transport via the roots and should be considered separately for the assessment of residues in  
370 floral matrices. In contrast to these findings, the tables provided by EFSA (2012, 2013) indicate that  
371 residue values from foliar applications are higher compared to soil treatments. However, those tables

372 only consider seed dressings, which contain significantly less active ingredient than soil drenches or  
373 foliar sprays. Furthermore, many residue values for foliar applications are derived from applications  
374 performed during bloom or shortly before, whereas the latest foliar application in Cowles' and Eitzer's  
375 experiment was applied two weeks before bloom.

376 Dively and Kamel (2012) showed that foliar-applied neonicotinoids in squash resulted in higher  
377 residues in pollen compared to a soil drip and drench application, especially when squash was sprayed  
378 at full bloom. In contrast, the PPP residues from foliar applications in nectar were lower after a spray  
379 application or did not differ from soil drench and drip irrigation. This leads to the assumption that  
380 systemic PPPs are provided over a longer period from the inside of the plant and thus have a greater  
381 probability to accumulate and express in nectar. Dively and Kamel found the lowest residue levels  
382 from imidacloprid bedding tray soil applications. This was the most distant application method relative  
383 to bloom and no increase in residues could be observed. However, the dose rate was very low  
384 compared to the other treatment regimes. In total, contrary to Cowles and Eitzer's (2017) experiment,  
385 the timing of the application and dose rate seemed to play a more important role than the application  
386 method, confirming the assumption that applications closer to bloom result in higher residues. Both  
387 Kubik et al. (1999) and Wallner (2009) showed that there is a lag period between the application of  
388 fungicides and the maximum residue level in pollen, although the compounds were sprayed directly  
389 onto the plant before and during bloom in cherry trees and OSR, respectively. More studies with  
390 different PPPs are necessary to confirm these results, especially for foliar applications (Figure 2).  
391 Overall, it can be concluded that there is a strong interdependence between the time available for the  
392 accumulation of the compound and the time for dissipation, metabolism and translocation in the  
393 plant, influenced by the chemical properties of a PPP and the application method.

#### 394 **Physicochemical Properties**

395 A detailed knowledge about the physical and chemical properties of a chemical compound is a  
396 necessary prerequisite to understand its general behaviour in metabolism, analytical methods,

397 formulations, and the environment (Tsipi et al. 2015). Therefore, these parameters are usually studied  
398 under well-defined conditions and are required for the registration of a PPP. Physicochemical  
399 properties determine the uptake of a compound into the plant, its translocation, as well as its  
400 dissipation and metabolisation in the plant and the environment.

401 For PPPs applied before bloom, it can be assumed that every parameter influencing the uptake of a  
402 compound and its acropetal translocation will influence the residues in floral resources. Some key  
403 physicochemical properties include the solubility in water, the partition coefficient octanol/water ( $\log$   
404  $K_{ow}$ ), the dissociation coefficient ( $pK_a$ ), the molecular size of a compound, the root concentration factor  
405 and the transpiration stream concentration factor. These properties can be altered by additives and  
406 vary depending on the formulation type (Bonmatin et al. 2015; Farha et al. 2016; Hsu and Kleier 1996;  
407 Trapp 2004).

408 Overall, PPPs can be classified according to their behaviour in and on plants. Non-systemic or contact  
409 compounds are not distributed in the plant and will probably cause only residues in floral matrices if  
410 the flower or pollen comes directly into contact with the PPP. On the contrary, translaminar PPPs are  
411 taken up and redistributed from one face of a leaf to the opposite face of a leaf, an important  
412 parameter for many fungicides (Klittich et al. 2008), whereas systemic PPPs are distributed within the  
413 whole plant, either acropetally via the transpiration stream to older leaves in xylem or in both,  
414 acropetal and basipetal directions to new growth in the phloem sap. The most common way for the  
415 translocation of (non-ionised) plant systemic insecticides is the unidirectional acropetal translocation  
416 in xylem (Wyss and Bolsinger 1997).

417 One key parameter describing PPP translocation for non-ionised compounds in the plant is the  
418 partition coefficient octanol/water ( $\log K_{ow}$ ). It describes the compound's lipophilicity and its ability to  
419 move through bio membranes; thus it determines the uptake of a PPP through the leaf cuticle and its  
420 distribution within the plant (Briggs and Bromilow 1994; Kirkwood 1999; Klittich et al. 2008; Wang and  
421 Liu 2007).

422 In general, compounds with a  $\log K_{ow} < 0$  are considered to be hydrophilic and compounds with a  $\log$   
423  $K_{ow} > 0$  lipophilic (Wang and Liu 2007). Lipophilic compounds tend to cross bio membranes but are  
424 partitioned into lipophilic tissue along the symplastic pathway (Sicbaldi et al. 1997). Therefore, the  
425 optimum uptake and translocation in xylem occurs for non-ionised PPPs with intermediate  $\log K_{ow}$   
426 values of 1–3. Translaminar distributed compounds can show  $\log K_{ow}$  values up to 4.5. Highly polar and  
427 highly non-polar compounds are poorly translocated within a plant (Bromilow and Chamberlain 1989;  
428 Bromilow and Chamberlain 1995; Sicbaldi et al. 1997; Vryzas 2016).

429 Non-ionised compounds with a lower  $\log K_{ow}$  can also be distributed in the phloem sap, though  
430 entering the symplast is impeded (Bromilow et al. 1987). In contrast, more lipophilic compounds can  
431 readily enter the phloem, but also easily move between xylem and phloem. However, as the xylem is  
432 moving faster than the phloem, compounds are eventually translocated in the xylem (Peterson and  
433 Edgington 1976; Wyss and Bolsinger 1997).

434 In general, most phloem-mobile compounds appear to be weak acids (Trapp 2004) and their  
435 translocation is highly dependent on a favourable combination of  $\log K_{ow}$  and the dissociation  
436 coefficient ( $pK_a$ ) (Wyss and Bolsinger 1997). The  $pK_a$  describes the acid strength and ability of a  
437 compound to dissociate; it can be regarded as the pH at which a particular acid or base group is 50%  
438 ionised (Bromilow and Chamberlain 1995). Plant compartments exhibit different pH values across  
439 membranes, ranging from pH 5 in the apoplast to pH 8 in the phloem sap (Chamberlain et al. 1998).

440 Accordingly, a weak acid will appear at low pH in its un-dissociated state, having the ability to easily  
441 enter the symplast. Once in the symplast, due to the higher pH, it dissociates and is not able to cross  
442 back through the membranes (i.e. the ion trap theory) (Briggs et al. 1987; Bromilow and Chamberlain  
443 1995; Chamberlain et al. 1998; Pessaraki 2014; Tyree et al. 1979).

444 It is understood that pollen and nectar, as part of reproductive organs, are a sink for photosynthetic  
445 products, even though nectaries can be supplied by phloem and xylem depending on the crop and  
446 variety (Heil 2011; Pacini et al. 2016; Wist and Davis 2006). However, many PPPs already found in  
447 these matrices, mainly insecticides and fungicides, are considered to be more xylem-mobile according

448 to their physicochemical properties. Thus, an acropetal movement in the plant is conceivable but the  
449 exact mechanism as to how these chemicals are transferred into the pollen is not yet understood.  
450 Aajoud et al. (2008) demonstrated that the low transpiration stream of different parts of a sunflower  
451 head cannot be responsible for all of the fipronil residues found in this tissue. Although fipronil is more  
452 likely to move in xylem due to its high  $\log K_{ow}$  (= 4.0), Aajoud et al. showed under laboratory conditions  
453 that fipronil is transported from sources (older leaves) to sinks (growing parts). In general, for non-  
454 ionised compounds like fipronil or neonicotinoids the ion trap theory does not apply and the active  
455 ingredient can move freely between phloem and xylem according to its bio membrane permeability  
456 (Sur and Stork 2003). Aajoud et al. (2008) suggested that both xylem and phloem pathways are  
457 involved in the transfer of fipronil to the flower head. Transfer via xylem from the roots to the leaves  
458 has been previously demonstrated and depends upon the rate of leaf transpiration, in addition to the  
459 compound concentration in the xylem, whereas the phloem pathway seems to be an influencing factor  
460 in the translocation to the flower parts and hence to pollen or nectar.

461 Another explanation for unexpected phloem transport is that biotransformation in plants can alter  
462 the compounds' properties. For example, due to its physicochemical parameters, imidacloprid is  
463 transported in xylem and accumulates in leaves, but some of its metabolites (e.g. 6-chloronicotinic  
464 acid) were shown to have properties which are potentially phloem-mobile (Buchholz and Nauen 2002;  
465 Chamberlain et al. 1995; Nauen et al. 1999). Furthermore, transformed compounds can form  
466 conjugates with glucose, isomaltose and amino acids, which could change the translocation pathway  
467 (Jiang et al. 2009; Oliver and Hewitt 2014; Sur and Stork 2003; Wu et al. 2012).

468 These findings could perhaps explain the increase in imidacloprid in upper plant parts, as described in  
469 the earlier section about crop-related parameters, and rationalise the presence of PPP residues in  
470 physiological sinks like pollen and nectar. Nevertheless, these conclusions might not apply for  
471 compounds with other physicochemical properties, especially for PPPs which are considered to be  
472 non-systemic.

473 In pollen residue studies, physicochemical properties are often mentioned to describe and explain the  
474 reason for differences in residue levels but, to our knowledge, no study has tried to link these PPP  
475 characteristics experimentally to the residues found in pollen or other matrices. Kyriakopoulou et al.  
476 (2017) found weak correlations between the residue levels in nectar and the solubility in water,  
477 although Bromilow and Chamberlain (1995) considered water solubility as a rather poor guide to  
478 systemic behaviour. Thorbek and Hyder (2006) used artificial neural networks to examine the  
479 relationship between physicochemical properties of different PPPs and residues in food products. In  
480 their opinion, the physicochemical properties and the crop type explained up to 50% of the variation.  
481 Thorbek and Hyder concluded that these properties control important aspects of the processes  
482 leading to residues in food commodities. These findings may be transferred to the residue occurrence  
483 in bee-important plant matrices.

484 In general, the uptake of PPPs and their half lives in plants are very well studied, primarily because risk  
485 assessments on human exposure or their environmental fate are required for the registration of PPPs,  
486 as well as the setting of Maximum Residue Levels (MRLs). However, the process determining the  
487 residues in bee-important matrices is not well understood, and more research is required to link the  
488 physicochemical properties of a compound to the translocation to pollen or nectar and to the fate and  
489 dissipation after the application of a PPP (Figure 3).

#### 490 **Environmental conditions**

491 PPPs applied to a crop enter a complex system, which is greatly influenced by its surrounding  
492 environment and underlying manifold interactions. These variations are reflected in the fluctuating  
493 PPP residues reported in pollen or nectar, especially in field experiments. Laurent and Rathahao (2003)  
494 reported significantly higher variations in pollen residues in a lysimeter experiment compared to  
495 greenhouse experiments. Jiang et al. (2018) experienced varying residue fluctuations across the  
496 course of one month in field experiments and Rolke et al. (2016) observed high variations even within  
497 different sub-areas of one field. Even small-scale weather incidents can change the result of a chemical

498 treatment, for example when the compound is washed off the leaves by rain shortly after application.  
499 Additionally, plant growth and physiology are dependent on the surrounding conditions and will  
500 influence the behaviour of the compound. This of course makes the comparability of PPP residues in  
501 pollen or nectar from different studies difficult, although the environmental fate of all kinds of PPPs  
502 are well studied.

503 Key parameters influencing both the chemical fate and the plant are water and temperature.  
504 Physicochemical properties are usually assessed under defined laboratory conditions and are  
505 therefore likely to change under varying conditions (Hornsby et al. 1995; Tsipi et al. 2015). For  
506 example, cuticle permeability was shown to increase rapidly with increasing temperatures (Baur et al.  
507 1997; Baur and Schönherr 1995). Degradation processes in soil, vegetation and air are all accelerated  
508 at higher temperatures (Bloomfield et al. 2006), whereas colder temperatures limit biological and  
509 chemical reaction activities, resulting in longer half-lives and slower dissipation rates (Farha et al.  
510 2016). Humidity can increase compound uptake into leaves (Hull 1970; Ramsey et al. 2002), while rain  
511 can lead to wash-off and leaching (Hunsche et al. 2007; Radolinski et al. 2018) and water stress was  
512 shown to affect the distribution of systemic insecticides in plant leaves (Stamm et al. 2016). Soil  
513 conditions are affected by temperature and water availability; organic matter content, microbial life  
514 and clay content play a key role in the fate and uptake of soil applied PPPs (Cessna et al. 2017; Di et  
515 al. 1998; Gevao et al. 2000; Zhang et al. 2018). PPPs with a long half-life in soil or exposed to conditions  
516 that prevent a breakdown in soil are more likely to be taken up during flowering. The transport of  
517 xylem-mobile compounds is, inter alia, dependent on the transpiration stream. Therefore,  
518 environmental conditions and plant species which enable a high transpiration will enhance acropetal  
519 movement and consequently the likelihood of translocation of PPP residues to pollen or nectar.

520 In general, flowers are also exposed to these conditions, however, they may show a different  
521 susceptibility to environmental conditions and a different uptake compared to the rest of the plant,  
522 due to the different structure and often hydrophilic properties of their surface (Baker and Hunt 1981;

523 Koch et al. 2008). Furthermore, flower opening, the dispersal of pollen, as well as the amount and  
524 composition of pollen and nectar produced is dependent on the surrounding environment, especially  
525 temperature and humidity (Heil 2011; Pacini et al. 2006; Vidal et al. 2006). PPPs applied during or  
526 shortly before bloom will contact the flowers directly and the presence of residues is therefore likely  
527 at least in pollen, even for compounds with a short half-life. All factors favouring a fast dissipation or  
528 degradation can thus decrease the residues in pollen or nectar. Choudhary and Sharma (2008)  
529 recorded a general faster dissipation of PPP residues in pollen than in nectar depending on the active  
530 ingredient. They attributed this faster degradation to the fact that pollen is more exposed to  
531 environmental conditions than the nectaries, which are typically deeply embedded within the flower.  
532 Consequently, the presentation of pollen, the arrangement of anthers and nectaries within the flower  
533 and their protection by flower petals could influence the impact of environmental conditions on  
534 residue behaviour in pollen and nectar. For example, compounds with a relatively low photo stability,  
535 such as pyrethroids, might dissipate faster in pollen grains presented openly to pollinators, compared  
536 to residues in nectar. None of the available studies considered or compared the influence of  
537 environmental conditions on PPP residues in pollen or nectar (Fig. 4). However, a field study conducted  
538 in consecutive years detected correlations in PPP residues from one year to another, despite varying  
539 environmental conditions (Dively and Kamel 2012). Nevertheless, the factors acting in different  
540 environments on PPPs availability in floral resources are complex and not well understood. [Different  
541 climates and soils, for example across Europe, are currently accounted for in risk assessments for bees  
542 by conducting residue trials at multiple sites across broad geographic regions. However,  
543 environmental influences are not understood well enough to allow an extrapolation or comparison  
544 between different sites and may require further attention depending on the mode of application and  
545 properties of the active ingredient \(e.g. soil uses with systemic compounds, UV stability\). Controlled-  
546 environment studies looking at the effect of for example temperature on residues could provide  
547 further insights.](#)

548



549 **Conclusions**

550 Overall, PPP residues in bee-important plant matrices are subject to manifold influences and many  
551 parameters can potentially impact their level and residence time in pollen or nectar. Several plant-  
552 related parameters, such as species and variety (including morphology), habitus and structure, were  
553 identified as contributing factors to the variation observed in PPP residue levels in pollen or nectar.  
554 Furthermore, the application mode, especially the dose rate and the timing of the application, were  
555 considered as a key source of residue variations. Nevertheless, the highest variations can probably be  
556 explained by the physicochemical properties of different compounds and, above all, by the influence  
557 of environmental conditions. However, we also demonstrate that studies which focus on these  
558 influencing factors are scarce and the complex processes which determine the residue level in bee-  
559 important matrices are not well understood (Fig. 1-4).

560 Thus far, research has typically concentrated on the influences of the broad application areas of  
561 neonicotinoids, thereby mainly on soil applications, which are not representative of most other  
562 insecticides. Investigations into the variability of non-systemic products in floral resources is notably  
563 neglected in research, whilst further research into residues of fungicides and herbicides in pollen and  
564 nectar is also required.

565 It is questionable whether the currently available data sets on residue levels can mitigate the  
566 described variability and whether they are representative enough to be used for conducting reliable  
567 risk assessments on pollinators. More wide-ranging and well replicated studies, which reflect different  
568 cropping scenarios, are necessary to obtain reliable residue levels in these specific matrices. In  
569 addition, PPPs are designed to have the best possible uptake rate and retention time on and in the  
570 plant to be effective against pests and to simultaneously avoid environmental pollution. This conflicts  
571 with the aim to achieve low residues in pollen or nectar. Therefore, application modes and  
572 circumstances in which PPP residues in flower parts are low or dissipate fast should be clarified.

573 It would be extremely difficult to assess the fate in pollen or nectar for all active ingredients, in all bee-  
574 important plants and under different climates. Therefore, methods are needed which enable an  
575 accurate estimation and extrapolation of PPP residues in these ecologically important matrices, which  
576 are also able to consider the numerous influences they are exposed to. In order to enable this,  
577 comparable results are required, which do not just reflect a snapshot of a single randomly selected  
578 field area and environmental conditions, but also reveal a broader knowledge which can be  
579 transferred to further situations.

580 This can only be achieved by improving the understanding of residue behaviour and their dynamics in  
581 these complex tissues. A fundamental challenge for future research will be to quantify the effects of  
582 different dynamics and interacting factors on PPP residue levels. Future research should aim to  
583 investigate relationships, interdependences and common features amongst various PPP applications,  
584 which may allow conclusions to be drawn on residues in pollen and nectar and, as a result, permit  
585 suitable systems to be identified which can act as model scenarios or be consulted for worst-case  
586 estimations, enabling all other scenarios to be adequately covered.

587 Achievement of this will permit risk assessments to be conducted with considerably less effort and  
588 expenditure, whilst simultaneously enabling rapid and accurate assessment of the risks for pollinators  
589 posed by PPPs.

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#### 594 **Conflict of interest**

595 We wish to draw attention to the fact that the corresponding author's PhD project is funded by  
596 Syngenta Ltd, a company manufacturing and selling plant protection products. Furthermore, some of  
597 the co-authors are employed by Syngenta.

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934 **Tables**

935 **Table 1** Summary of selected PPP residues in pollen expressed as Residue Unit Dose (RUD) (mg a.i. kg<sup>-1</sup> pollen at an  
 936 application rate of 1 kg a.i. ha<sup>-1</sup>) derived from two different sources. The minimum, maximum and mean values demonstrate  
 937 the high variability of residues found in pollen from spray applications. Some calculations were not derived from a single  
 938 crop or active ingredient, but many different crops/active ingredients were summarised (“various”).

Crop	Active ingredient	Min (RUD mg kg <sup>-1</sup> )	Max (RUD mg kg <sup>-1</sup> )	Mean (RUD mg kg <sup>-1</sup> )	Source*
various	various	0.0002	149.8	6.1 ± 30.704 (SD)	a
various	various	0.004	366	65.06 ± 89.421 (SD)	b
various	alpha- Cypermethrin	11.370	366.3	167.3 ± 121.438 (SD)	b
<i>Brassica</i> sp.	various	2.083	366.3	87.06 ± 102.8 (SD)	b
	Teflubenzuron	21.7	149.8	**	a
	Acetamiprid	3.4	14.8	**	a

**Examples for PPP residues of the same active ingredient in different crops**

Active ingredient	Crop			Source*
Spirotetramat	Melon ( <i>Cucurbitaceae</i> )	<i>Phacelia tanacetifolia</i> L.	<i>Brassica</i> sp.	a
	2.2	63.5	83.1	

939 \*Sources: a) EFSA 2013, see Annex F; b) Kyriakopoulou et al. 2017

940 \*\* Only two residue values were provided for this active ingredient/ crop combination

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945 **Table 2:** Relationship between dose rate, residues in pollen (ppm) and Residue Unit Dose (RUD) for three active

946 ingredients. Data from Choudhary and Sharma (2007). Application of 750 L ha<sup>-1</sup> water in mustard (*Brassica juncea* Czern.)

947 in 2003/2004.

Active ingredient	Dose rate (g a.i. ha <sup>-1</sup> )	Residues pollen ppm	RUD
Endosulfan	525	2.126	4.05
Spiromesifen	225	2.052	9.12
Lambda- Cyhalothrin	75	1.607	21.43

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