Addressing bystander exposure to agricultural pesticides in life cycle impact assessment

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1 **Addressing bystander exposure to agricultural pesticides in life cycle impact**

2 **assessment**

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Abstract

Residents living near agricultural fields may be exposed to pesticides drifting from the fields after application to different field crops. To address this currently missing exposure pathway in life cycle assessment (LCA), we developed a modelling framework for quantifying exposure of bystanders to pesticide spray drift from agricultural fields. Our framework consists of three parts addressing: (1) loss of pesticides from an agricultural field via spray drift; (2) environmental fate of pesticide in air outside of the treated field; and (3) exposure of bystanders to pesticides via inhalation. A comparison with measured data in a case study on pesticides applied to potato fields shows that our model gives good predictions of pesticide air concentrations. We compared our bystander exposure estimates with pathways currently included in LCA, namely aggregated inhalation and ingestion exposure mediated via the environment for the general population, and general population exposure via ingestion of pesticide residues in consumed food crops. The results show that exposure of bystanders is limited relative to total population exposure from ingestion of pesticide residues in crops, but that the exposure magnitude of individual bystanders can be substantially larger than the exposure of populations not living in the proximity to agricultural fields. Our framework for assessing bystander exposure to pesticide applications closes a relevant gap in the exposure assessment included in LCA for agricultural pesticides. This inclusion aids decision-making based on LCA as previously restricted knowledge about exposure of bystanders can now be taken into account.

1.   Introduction

Residents living within short distance of agricultural fields may be exposed to spray drift emissions from pesticides designed to target field pests and weeds, during and after applications (Matthews and Hamey, 2003). Methods for estimating spray drift and potentially related exposure of residential bystanders have been proposed based on either empirical measurements (Ganzelmeier et al., 1995; Holterman and van de Zande, 2003; Kasiotis et al., 2014; Rautmann et al., 2001; Salyani and Cromwell, 1992; van de Zande et al., 2010) or mechanistic models (Arya, 2003; Craig, 2004; Kennedy et al., 2012; Lebeau et al., 2011; Miller and Hadfield, 1989; Reiss and Griffin, 2006; Teske et al., 1993, 2002). A detailed overview of identified spray drift models is provided in Section S-5 of the Supplementary material. These models are usually applied to support environmental risk assessment (ERA), while estimating pesticide exposure associated with spray drift fractions for bystanders living near agricultural fields is currently not included when comparing agricultural practices in a life cycle assessment (LCA) context (Rosenbaum et al., 2015). This is mainly due to the fact that while most of the existing modeling approaches are suitable for quantifying drift-related deposition profiles, their specific environmental and boundary conditions and underlying assumptions are usually not appropriate for assessing residential bystander exposure in an LCA context. LCA requires considering the number of residential bystanders in a defined area that are exposed to the integrated drift-related pesticide emission amount reaching that area from a pesticide application to the total agricultural field. Such a modeling approach, however, is currently not available for LCA. Hence, the potential contribution of exposure associated with spray drift fractions to overall population exposure and related impacts on human health relative to other exposure pathways including exposure to crop residues (Fantke et al., 2012a; Fantke and Jolliet, 2016) is currently unknown. This may lead to uninformed decisions and in some cases shift burden from population far-field or crop residue exposure to exposure of bystanders.

61   To address this issue, we aim in the present study to develop and apply a framework for

62   quantifying exposure of bystanders to pesticide spray drift fractions from agricultural fields for

63   integration in life cycle impact assessment (LCIA), and focus on the following specific objectives:

64   (1) to propose a modeling framework for estimating bystander inhalation exposure to agricultural

65   pesticides from spray drift, (2) to evaluate the model in terms of uncertainty using Monte Carlo

66   simulation and in terms of comparing modeled air concentrations with field measurements in a case

67   study of pesticides applied to potato fields, and (3) to compare the magnitude of estimated

68   bystander exposure with overall population exposure to environmental far-field emissions (via

69   fractions lost to the environment during and after pesticide application) and population exposure to

70   residues in food crops. The results of this study close a known gap in exposure modelling in LCIA

71   toxicity characterization by providing a quantitative estimate of inhalation exposure of bystanders to

72   agricultural pesticides.

73

74   **2.   Materials and Methods**

75   **2.1.   Model framework**

76   We propose a model to estimate inhalation exposure of bystanders living in the vicinity of

77   agricultural fields to the integrated air-borne pesticide spray fraction drifting beyond the field

78   boundaries during and after application for inclusion as complementary exposure pathway in LCIA

79   human toxicity characterization. A conceptual overview of the model setup is presented in Figure 1.

80   In the following, we provide an overall description of the modeling framework, while further details

81   of the approach and underlying model equations can be found in Section S-1 of the Supplementary

82   material.
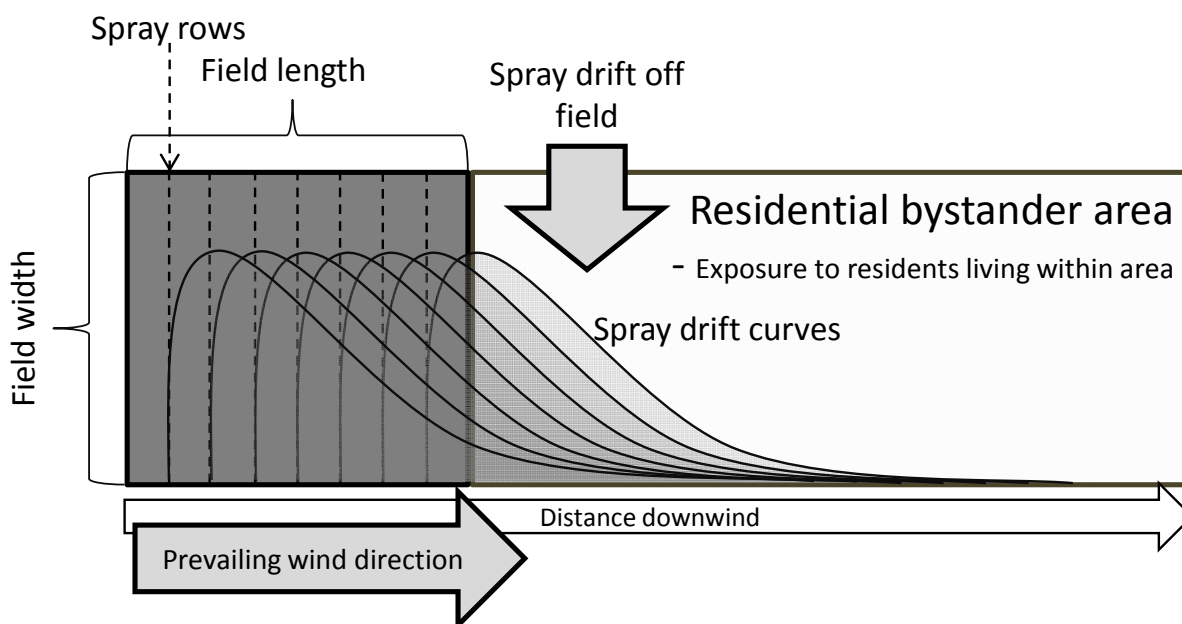
83

84   Figure 1 Conceptual overview of the approach to model pesticide spray drift from a field, and the

85   model boundaries, where subsequent exposure to the integrated spray drift fraction over the

86   considered residential bystander area is included. Off-field spray drift is inversely correlated with

87   the distance between the sprayed field row and the field edge.

88

89   The residential bystander exposure model consists of three main parts: (1) the loss of

90   pesticide from the agricultural field via spray drift after application; (2) environmental fate of the

91   pesticides in air outside of the treated field; and (3) exposure of the bystander population to the

92   pesticide spray drift fraction. Spray drift loss and environmental fate processes are considered by

93   the fate factor, FF $[\mathrm{kg_{in\,air}}$ per $\mathrm{kg_{applied}\,d^{-1}}]$, while human exposure is represented by the exposure

94   factor, XF $[\mathrm{kg_{inhaled}\,d^{-1}}$ per $\mathrm{kg_{in\,air}}]$, integrated over the boundaries of the considered bystander

95   residence area. The human intake fraction iF $[\mathrm{kg_{inhaled}\,kg_{applied}^{-1}}]$ is a representative metric to

96   express the mass of pesticide inhaled by the entire bystander population per unit of mass applied

97   pesticide and can be calculated as the product of fate and exposure, i.e. FF×XF. The iF can then be

98   multiplied with a human toxicity-related effect factor, EF $[\mathrm{disease\,cases\,kg_{inhaled}^{-1}}]$ that relates

99   human inhalation intake to disease risk (Crettaz et al., 2002; Jolliet et al., 2006) to finally arrive at a

100   characterization factor, CF [disease cases $\mathrm{kg}_{\mathrm{applied}}^{-1}$], expressing the potential impacts on human

101   health of bystanders from agricultural pesticide applications:

102     $\mathrm{CF} = \mathrm{FF} \times \mathrm{XF} \times \mathrm{EF} = \mathrm{iF} \times \mathrm{EF}$ Eq. 1

103   The residential bystander model estimates the exposure of residential and non-residential bystanders

104   from initial spray drift. Exposure associated with continuous processes, such as volatilization and

105   subsequent inhalation or deposition onto field crops and subsequent ingestion of crop residues, are

106   not considered in the presented model, as the contribution of these pathways to overall population

107   exposure is already considered by existing LCIA models, such as USEtox (Rosenbaum et al., 2008)

108   and dynamiCROP (Fantke et al., 2011a, 2011b).

109

110   **2.1.1   Environmental fate of pesticides applied to fields**

111   The fate of the pesticides as expressed by the FF was estimated according to:

112     $\mathrm{FF} = \dfrac{m_{\mathrm{in\,air}}}{\dot{m}_{\mathrm{applied}}} = \dfrac{\sum_i m_{\mathrm{in\,air},i}}{\sum_i \dot{m}_{\mathrm{applied},i}}$ Eq. 2

113   where $m_{\mathrm{in\,air},i}$ [$\mathrm{kg}_{\mathrm{in\,air}}$] is the time-integrated mass in air associated with spray row i, and $\dot{m}_{\mathrm{applied},i}$

114   [$\mathrm{kg}_{\mathrm{applied}}\,\mathrm{d}^{-1}$] is the mass applied per day to spray row i. The mass found in air is essentially a

115   function of two components (see Supplementary material, Eq. S6) in line with recommendations

116   based on a broad agreement regarding the modeling of the delineation of pesticide environmental

117   distribution processes between life cycle inventory (LCI) and LCIA (Rosenbaum et al., 2015). The

118   first component is considered in the LCI phase estimating the fraction of the pesticide applied to the

119   field that is lost via spray drift, while the second component is considered in the LCIA phase

120   estimating the fate of pesticides in air outside of the treated field.

121   Losses of pesticides from agricultural fields have been extensively assessed, where focus has

122   primarily been on the deposition of the pesticides (Ganzelmeier et al., 1995; Holterman and van de

123   Zande, 2003; Teske et al., 2002). Based on these studies, we calculated spray drift as a fraction of

124   the applied pesticide mass separately for each nozzle row i according to available drift deposition

125   curves by e.g. Holterman and van de Zande (2003), which estimate the deposited fraction as a

126   function of distance from the application site. At sufficiently large distance from the place of

127   application, the entire fraction drifted from the field was assumed deposited. The deposition curves

128   do not include the spray drift fraction that remains airborne, thus, slightly underestimating the total

129   losses from the field. Nevertheless, this method was regarded as a good proxy for the total loss via

130   spray drift. Based on this assumption, we estimated the applied pesticide fraction lost via wind drift

131   using an emission quantification model building on PestLCI 2.0 (Dijkman et al., 2012), which we

132   adapted to account for integrated emission mass beyond agricultural fields, buffer zones and drift

133   curves applicable for potato cultivation:

134   $$fr_{\text{drift},i} = \frac{1}{100} \times \left[\frac{\alpha_1}{\beta_1} \left(e^{-z_{1,i} \times \beta_1} - e^{-z_{2,i} \times \beta_1} \right) + \frac{\alpha_2}{\beta_2} \left(e^{-z_{1,i} \times \beta_2} - e^{-z_{2,i} \times \beta_2} \right) \right] \qquad \text{Eq. 3}$$

135   where $fr_{\text{drift},i}$ [$\text{kg}_{\text{drift}}\ \text{kg}_{\text{applied}}^{-1}$] is the fraction of pesticide lost from the field via spray drift and

136   reaching the exposed area per mass applied in nozzle row $i \in \{1, ..., n\}$, $z_{1,i}$ [m] is the distance of

137   spray row i to the field edge, $z_{2,i}$ [m] is the distance of spray row i to field edge plus 10,000 m after

138   which all spray drift is assumed to be deposited, and $\alpha_1, \alpha_2, \beta_1, \beta_2$ are regression coefficients,

139   which primarily depend on crop type, nozzle type and application technique (Holterman and van de

140   Zande, 2003). The implemented drift deposition curves are based on spray drift of pesticides in

141   aqueous formulations, for which our model is valid. Implementation of other spray drift curves (e.g.

142   from Ganzelmeier et al., 1995; Kasiotis et al., 2014; Rautmann et al., 2001) is possible by adapting

143   the integrated function in Eq. 3 to be based on the equation specific to any new spray drift curve.

144   Drift curves for similar crops and similar application techniques were found to vary substantially

145   between studies. For instance, the total drift-related loss of pesticide from a single spray row

146   expressed as % of applied mass to potato crops using conventional ground boom sprayers was

147   found to range from 15% to 73% depending on the spray drift curves used (see Supplementary

7

148 material, Table S10). This variation is likely a result of differences in spray technologies used in the

149 underlying spray drift studies, in environmental conditions during measurements, and in

150 measurement setups and technologies.

151   For pesticide emissions beyond agricultural fields from each nozzle row i, the mass fraction

152 of pesticides in the air was estimated as a function of distance based on a Gaussian plume

153 dispersion model including terms for advection, horizontal and vertical dispersion processes

154 yielding a time-integrated estimate of the average air mass fraction in the exposed bystander area as

155 a function of distance:

156

157
$$fr_{\mathrm{disp},i} = \underbrace{\frac{1}{c_{\mathrm{s/d}}} \times \frac{1}{2 \times \pi \times u \times \sigma_y \times \sigma_z}}_{\text{advection}} \times \underbrace{e^{-\frac{1}{2}\times\left(\frac{y}{\sigma_y}\right)^2}}_{\text{horizontal dispersion}} \times fr_{\mathrm{size}} \times \underbrace{\left[e^{-\frac{1}{2}\times\left(\frac{h_{\mathrm{app}}+x\times\frac{v}{u}}{\sigma_z}\right)^2} + fr_{\mathrm{reflect}} \times e^{-\frac{1}{2}\times\left(\frac{h_{\mathrm{app}}-x\times\frac{v}{u}}{\sigma_z}\right)^2} \right]}_{\text{vertical dispersion}} \quad \text{Eq. 4}$$

158 where $fr_{\mathrm{disp},i}$ [$kg_{\mathrm{dispersed}} \, kg_{\mathrm{drift}}^{-1}$] is the mass fraction of spray drift lost from the field from

159 application in row i and dispersed in downwind direction at distance x within the exposed area,

160 $c_{\mathrm{s/d}}$ [$s \, d^{-1}$] is a conversion factor of seconds per day to correct for the unit of inputs to the plume

161 model, x [m] is the downwind distance between field edge and point distance in the exposed

162 bystander area, y [m] is the transversal distance away from the center of the plume, u [$m \, s^{-1}$] is the

163 average wind velocity, σ_y and σ_z [m] are horizontal and vertical dispersion coefficients of the

164 spray cloud, respectively (US Environmental Protection Agency, 1993), h_{app} [m] is the height of

165 spray application which is application technique and crop specific. A default h_{app} of 0.5 m above

166 crop canopy for potatoes was used based on Holterman and van de Zande (2003). v [$m \, s^{-1}$] is the

167 settling velocity of spray droplets, fr_{size} [$kg_{\mathrm{drop\text{-}size}} \, kg_{\mathrm{total}}^{-1}$] is the mass fraction of the total source

168 strength in the selected drop-size category, and fr_{reflect} [$kg_{\mathrm{reflected}} \, kg_{\mathrm{dispersed}}^{-1}$] is the mass fraction

169 that is reflected (re-emitted) at deposition surfaces. We use a constant prevailing wind direction

170 under the assumption that an average agricultural field is equally surrounded by residential

171   bystander areas in all directions. Thereby, the population-level exposure from a change in wind

172   direction would be the same, which is what we are after in a LCA context where the specific

173   location of a treated field is usually unknown. Further details of the emission and fate calculation

174   are given in Eqs. S1-S10 of the Supplementary material.

175

176   **2.1.2   Residential bystander population exposure**

177   Human exposure via inhalation of residential bystanders living in any pre-defined, exposed

178   area outside a treated agricultural field can be described by an exposure factor:

179    $$\text{XF} = \frac{\text{IR} \times n_{\text{pop}}}{V_{\text{air}}} \times c_{\text{exposure}}$$ Eq. 5

180   where XF $[\text{kg}_{\text{inhaled}}\, \text{d}^{-1}\, \text{per kg}_{\text{in air}}]$ is the bystander inhalation exposure factor within the exposed

181   area, IR $[\text{m}^3_{\text{inhaled}}\, \text{capita}^{-1}\, \text{d}^{-1}]$ is the average individual human inhalation rate, n_{pop} [capita] is the

182   number of bystanders living in the exposed area, V_{air} $[\text{m}^3_{\text{air}}]$ is the volume of air in the exposed area,

183   and c_{exposure} is a factor correcting for the fact that bystanders during the spray drift period spend

184   part of the time outdoors and part of the time indoors, both within the exposed area (US

185   Environmental Protection Agency, 2011).

186   Defining the size of the exposed area is essential for modeling exposure of bystanders as it

187   influences the exposed bystander population and related air volume. The width of the exposed area

188   was considered equal to the width of the treated agricultural field (w_{field}; m). The field width was

189   based on an estimation of typical European field sizes, which were between 8 to 20 hectares with

190   lengths between 600 and 700 m (European Commision - Joint Research Centre, 2012). This gives a

191   typical field width between 150 and 330 m, from which we defined a default field width of 300 m

192   for the present study. The length of the exposed bystander area outside the field (*l*; m*)* was defined

193   by three archetype distances (i.e. 8 m, 50 m and 100 m downwind) within which the exposed

194   residents are assumed to be located. The three distances used were based on distances typically

9

195 evaluated also in risk assessment (Garreyn et al., 2003; Kruijne et al., 2011; Martin et al., 2008) and

196 allow for evaluating exposure within different area sizes.

197   As the XF is directly proportional to the exposed population size, the number of inhabitants

198 living in the area exposed to pesticide spray drift is an important parameter. The population size in

199 the exposed area was calculated as:

200 $n_{\mathrm{pop}} = \rho_{\mathrm{pop}} \times w_{\mathrm{field}} \times l$ Eq. 6

201 Where ρ_{pop} is the population density in the exposed bystander area [capita $\mathrm{m}^{-2}_{\mathrm{exposed}}$]. Specific data on

202 population density near agricultural fields were not available and would likely also show large

203 spatial variability. Therefore, the population density as used in USEtox for the continental scale (i.e.

204 111 capita per km^2 for an average, generic continent (Rosenbaum et al., 2011)) was used as a

205 default value. The sensitivity of the model to this parameter was tested as part of the sensitivity and

206 uncertainty analysis. Further details of the exposure factor calculation and underlying terms are

207 given in Eqs. S11-S13 of the Supplementary material, and information on how we corrected for

208 differences between outdoor and indoor air concentrations are provided in Section S-3 of the

209 Supplementary material.

210

211 **2.2.   Model evaluation**

212 **2.2.1   Model sensitivity and uncertainty analysis**

213   A Monte Carlo simulation was conducted to determine the probability distribution and

214 variability of the model results and to evaluate the robustness of the model results toward changes

215 in model inputs. Input parameters for the residential bystander model were assigned a probability

216 distribution and probability parameters (see Supplementary materials, Table S8), and the Monte

217 Carlo simulation was run for 10,000 iterations to estimate the probability distribution of the intake

218 fraction, iF, for the residential bystander population at four distance intervals, namely 0 to 100 m, 0

219 to 8 m, 8 to 50 m, and 50 to 100 m from the edge of the treated agricultural field.

220   Based on the Monte Carlo simulation, the squared geometric standard deviation (GSD^2) of

221   model output was estimated as:

222    $$GSD^2 = \sqrt{\frac{97.5\%\text{-ile}}{2.5\%\text{-ile}}}$$ Eq. 7

223   The GSD^2 describes the variance of the iF uncertainty range across a 95% confidence interval

224   assuming an ideal lognormal distribution (Fantke et al., 2012b), which is plausible for many

225   parameters involved in calculating environmental processes (Limpert et al., 2001; Ott, 1990).

226   To demonstrate how to compare uncertainty estimates of spray drift related characterization

227   factors, CFs, for residential bystanders to general population related CFs for impacts on human

228   health obtained with USEtox for far-field environmental exposure pathways, iF and related GSD^2

229   values were coupled with effect factors found in the USEtox organic substances database

230   (http://usetox.org) with data for both carcinogenic and non-carcinogenic effects. The uncertainty of

231   the iF was estimated from uncertainty propagation using Taylor series expansion (MacLeod et al.,

232   2002) coupled with the uncertainty of the EFs based on Huijbregts et al. (2005) to give the overall

233   GSD^2 of CFs for bystander exposure in the 0 to 100 m bystander exposure area. Further details on

234   the Monte Carlo simulation are provided in Section S-4 of the Supplementary material.

235

236   **2.2.2   Evaluation against measured data**

237   Regardless whether model output is or is not sensitive toward changes in model inputs, the

238   accuracy of the model in terms of predicting the "true" value may still be low due to the potential

239   influence of data variability and the relatively small number of model inputs (Huijbregts, 1998).

240   Hence, we also evaluated our bystander exposure model's predictive ability by comparing predicted

241   average air concentrations (which are the basis for the subsequent inhalation exposure of

242   bystanders) with measured air concentrations based on the relative difference between predicted and

243   measured air concentrations:

11

244 $\quad$ Relative difference $\left(C_{\text{predicted}}, C_{\text{measured}}\right) = \dfrac{\left|C_{\text{predicted}} - C_{\text{measured}}\right|}{C_{\text{measured}}}$ $\hspace{3cm}$ Eq. 8

245 $\quad$ where $C_{\text{predicted}}$ [µg m^{-3}] is the average air concentration over the full considered exposure range of

246 $\quad$ 0 to 100 m distance from the treated potato field edge estimated with the residential bystander

247 $\quad$ model, and C_{measured} [µg m^{-3}] is the measured air concentration averaged over 100 m from the field

248 $\quad$ edge within 24 hours after pesticide application based on reported data from a treated potato field in

249 $\quad$ Prince Edward Island, Canada (Garron et al., 2012, 2009). We adapted the measured data to reflect

250 $\quad$ air concentrations that are integrated over distance and time to enable comparison with results from

251 $\quad$ our residential bystander model. Our comparison of model results with measured data was restricted

252 $\quad$ to a very limited set of scenarios, where actual information on field size, application method and

253 $\quad$ meteorological conditions was reported as required input to the residential bystander model. This is

254 $\quad$ in line with recommended data reporting requirements using experimental data inputs to facilitate

255 $\quad$ the parameterization and evaluation of estimation models (Fantke et al., 2016).

256

257 $\quad$ **2.2.3 Evaluation against other exposure pathways**

258 $\quad$ We finally compared our iF estimates for bystanders between 0 and 100 m from the treated

259 $\quad$ field edge with iFs for other exposure pathways including aggregated inhalation and ingestion

260 $\quad$ exposure mediated via the environment for the general population estimated with the USEtox model

261 $\quad$ (Rosenbaum et al., 2011, 2008), and general population exposure via ingestion of pesticide residues

262 $\quad$ in consumed food crops estimated with the dynamiCROP model (Fantke et al., 2011a, 2011b).

263 $\quad$ The comparison is based on estimating iFs from USEtox and dynamiCROP and normalizing

264 $\quad$ USEtox iFs to exposure per kg applied to allow comparison of iFs estimated with the residential

265 $\quad$ bystander model and dynamiCROP. Thereby, the pesticide mass in the USEtox air compartment

266 $\quad$ was linked to the mass applied by introducing a generic fraction of the applied mass (spray drift

267 $\quad$ plus volatilization) lost to the air compartment. For pesticide spray applications to potatoes, this

268   fraction was estimated to be 4.85% with an additional 10% loss to account for initial volatilization

269   of pesticides (Fantke and Jolliet, 2016). For dynamiCROP, the exposed population was estimated

270   from the total harvested annual potato yield of 1.6 kg m^{-2} (Juraske et al., 2011) multiplied with the

271   considered field area from Table 1 and divided by the annual amount of potatoes consumed per

272   capita of 32 kg capita^{-1} (FAO, 2013). The comparison was done for carbofuran (insecticide),

273   chlorothalonil (fungicide).

274

275   **3.   Results**

276   **3.1.   Bystander exposure and comparison with measured data**

277   Figure 2 shows estimated average air concentrations integrated over all spray rows within

278   residential bystander areas over different distance ranges from the edge of a treated agricultural

279   field. It is apparent that concentrations are substantially higher close to the field edge and that

280   concentrations decrease rapidly after about 10 m from the field. The difference in average air

281   concentrations between the 0 (agricultural field edge) to 8 m bystander range and the 50 to 100 m

282   bystander range is about four orders of magnitude. Indeed, the average air concentration between 0

283   and 100 m was found to be dominated by the concentration within the initial 0 to 8 m range.
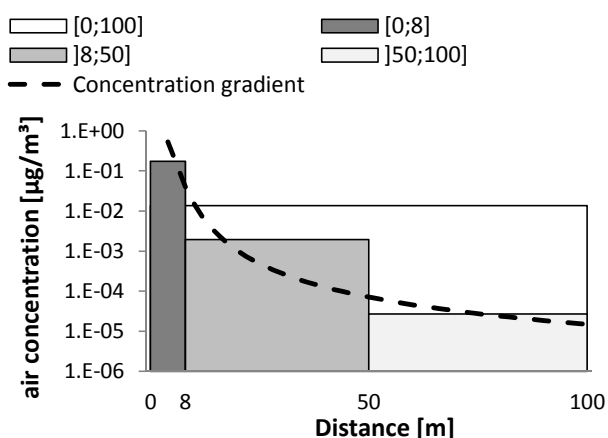
285   Figure 2 Gradient in outdoor air concentration integrated over all spray rows from 0 to 100 m range

286   as a result of 1 kg of pesticide, with chlorothalonil as active ingredient, applied to a generic

287   agricultural field using a conventional boom sprayer. The figure also shows the average outdoor air

288 concentration integrated over all spray rows within four distinct ranges over distances from the

289 treated agricultural field edge covering potentially exposed bystander areas.

290

291   Table 1 shows estimated iFs based on parameters reported together with air concentration

292 measurements (Garron et al., 2012, 2009). Across application scenarios of different pesticides and

293 different bystander exposure distance ranges, iFs differ by less than one order of magnitude within

294 the 0 to 8 m and 0 to 100 m, distance ranges (where the latter range is dominated by the 0 to 8 m

295 range), while the difference in iFs between scenarios was substantially larger with about four orders

296 of magnitude within the 8 to 50 m and 50 to 100 m distance ranges. Table 1 also shows how model

297 predictions for pesticide air concentrations match with reported field measurements. The relative

298 difference between the predicted (i.e. modeled) average air concentration between 0 and 100 m

299 exposed bystander range and the reported average air concentration between 0 and 100 m measured

300 over the first 24 hours after pesticide application to potato fields ranges between 0.07 and 1.47. We

301 observe a notable difference between measured and predicted air concentrations. However, the

302 difference is considered acceptable for providing a realistic indication of the time and space

303 integrated air concentration. This is also true when taking into account the inherent variability in

304 measured spray drift concentrations and the general uncertainty related to describing air

305 concentrations as a function of wind speed, temperature, relative humidity, etc. (see Supplementary

306 material, Section S2). Therefore, the modeling approach is considered of suitable accuracy for

307 providing a representative estimate of pesticide concentration in air from spray drift as a result of

308 pesticide application to potato fields.

309

310   Table 1 Predicted bystander population intake fractions (iF) for different pesticide

311 application scenarios and different exposure distance ranges from the treated agricultural field,

312 predicted ($C_{predicted}$) and measured ($C_{measured}$) air concentrations between 0 and 100 m and their

313 relative difference, and main model inputs, namely wind speed (u), relative humidity (rh), air

14

314   temperature (T_{air}), mass of pesticide applied (m_{app}), treated field area (A_{field}), field length along the

315   prevailing wind direction (L_{field}), application duration (t_{app}), and population density (ρ_{pop}). All

316   scenarios represent a separate field trial test, where pesticides have been applied and measured at

317   different distances from the field edge and at different times over the course of one day.

| Scenario		Model inputs								iF [$kg_{inhaled}$ $kg^{-1}_{applied}$]				$C_{predicted}$ [µg m^{-3}]	$C_{measured}$[3] [µg m^{-3}]	Relative difference
#	Pesticide	u [m s^{-1}]	rh [%]	T_{air} [°C]	m_{app} [kg]	A_{field} [ha]	L_{field} [m]	t_{app} [hr]	ρ_{pop} [capita m^{-2}]	0 to 8 m	8 to 50 m	50 to 100 m	0 to 100 m	0 to 100 m	0 to 100 m	
1[1]	Chlorothalonil	3.5	79	18.4	12.1	10.1	336	1.2	1.11×10^{-4}	5.9×10^{-10}	7.6×10^{-11}	2.7×10^{-12}	6.1×10^{-10}	0.22	0.11	1.05
2[1]	Chlorothalonil	2.1	69	19.9	12.1	10.1	333	0.6	1.11×10^{-4}	8.6×10^{-10}	6.5×10^{-12}	8.8×10^{-14}	7.9×10^{-10}	0.29	0.34	0.14
3[1]	Chlorothalonil	1.6	78	20.9	4.8	4	133	0.4	1.11×10^{-4}	2.5×10^{-9}	1.6×10^{-13}	7.3×10^{-16}	2.3×10^{-9}	0.33	0.36	0.07
4[1]	Chlorothalonil	2.1	73	26.3	4.8	4.0	133	0.4	1.11×10^{-4}	1.9×10^{-9}	1.3×10^{-11}	1.6×10^{-13}	1.8×10^{-9}	0.26	0.57	0.55
5[2]	Carbofuran	3.0	83	16.3	6.4	12.1	270	0.6	1.11×10^{-4}	1.1×10^{-9}	4.9×10^{-11}	1.2×10^{-12}	1.0×10^{-9}	0.13	0.08	0.61
6[2]	Methamidophos	2.7	68	15.6	8.7	8.1	403	0.5	1.11×10^{-4}	3.4×10^{-10}	1.5×10^{-11}	3.5×10^{-13}	3.3×10^{-10}	0.13	0.66	0.80
7[2]	Mancozeb	4.0	94	18.6	21.8	12.1	403	0.6	1.11×10^{-4}	3.2×10^{-10}	6.3×10^{-11}	2.7×10^{-12}	3.5×10^{-10}	0.23	0.09	1.47

318   [1]Data from Garron et al. (2012); [2]Data from Garron et al. (2009); [3]Experimental data were averaged from 0 to 24 hr and

319   from 0 to 100 m to get results that were comparable with outputs from the residential bystander model.

320

3.2.   Model sensitivity and uncertainty results

322   Based on the Monte Carlo simulation, Figure 3 shows the variability of bystander population

323   iF (i.e. intake fraction of all bystanders considered within the exposed area next to the treated

324   agricultural field) and individual bystander iF^{person} (i.e. intake fraction of a single bystander person

325   determined by dividing the exposure factor by the number of bystanders living in the considered

326   exposed area; see Eq. 5). The variability in iF values for the 0 to 100 m and 0 to 8 m distance ranges

327   is very similar, because iF for the 0 to 100 m range is again dominated by iF for the 0 to 8 m range.

328   The 95% confidence interval is about one order of magnitude for both total bystander iF and

329   individual bystander iF^{person} in the 0 to 100 m and 0 to 8 m distance ranges, which indicates

330   relatively little variability. The parameters contributing most to variability are wind speed,

331   application duration and individual inhalation rate for iF^{person}, while these parameters together with

332   bystander population density are dominating for total bystander iF. For the 8 to 50 m and 50 to 100

333   m distance ranges, the 95% confidence interval spans over three orders of magnitude and the

334   parameters contributing most to variability were wind speed, relative humidity and application

335   duration for iF^{person}, while these parameters together with population density are dominating for

336   total bystander iF. Details of the sensitivity of modeled air concentrations as output of the fate

337   model toward the aforementioned input parameters are provided in Section S-4 of the

338   Supplementary material. In general, little difference in variability was found at the exposure results

339   level between iF and iF^{person}, which is primarily due to the relatively low population density within

340   the exposed bystander area, where e.g. the total population living within the total 0 to 100 m

341   distance range is on average only 3.37 persons. Hence, the varying population within the bystander

342   area does not differ much from the iF^{person} where only a single bystander is considered.

343
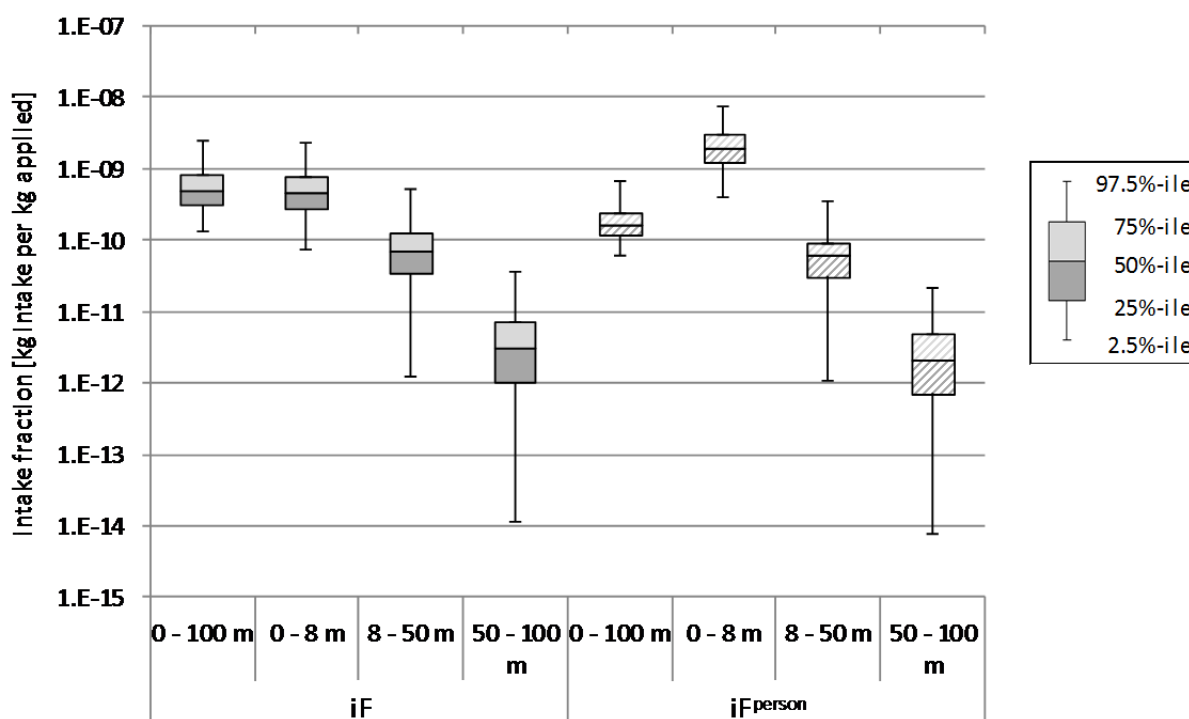
344

345   Figure 3 Box plots showing intake fraction ranges across Monte Carlo simulations for four

346   bystander exposure distance ranges from the field edge, highlighting the difference in exposure at

347   different distances to the treated field and also evaluating exposure per capita compared to exposure

348   of the entire considered bystander population.

349

350   Overall, our bystander exposure model does not appear highly sensitive to changes in model

351   inputs as the GSD^2 for iF$_{[0-100m]}$ was 4.27 and the corresponding lower and upper 95% confidence

16

352   interval limits are respectively 1.3×10^{-10} and 2.4×10^{-9} kg inhaled by the bystander population per

353   kg applied pesticide. This means that between 0.13 and 24 ppb (parts per billion) of applied

354   pesticides are inhaled by bystanders with 95% certainty in our scenarios. After coupling the

355   uncertainty of the bystander intake fraction in the range 0 to 100 m, $iF_{[0-100m]}$, with the uncertainty of

356   the effect factor, EF, linking exposure to disease risk, the GSD^2 of the corresponding

357   characterization factors, $CF_{[0-100m]}$, were 78.7 and 192.4 for non-carcinogenic and carcinogenic

358   health effects, respectively. Based on this scenario, the GSD^2 for $CF_{[0-100m]}$ lies within the

359   uncertainty for other CFs for impacts on human health ranging from 77 to 2189 depending on the

360   emission compartment and exposure pathway considered (Rosenbaum et al., 2008). However, the

361   highest contribution to the uncertainty of our bystander exposure related CFs is not associated with

362   our estimated intake fractions, but with the extrapolated human effect factor, especially for the

363   widely varying non-cancer effects, which is consistent with other studies (Fantke et al., 2012a;

364   Huijbregts et al., 2005; Rosenbaum et al., 2008).

365

3.3.   Comparison of different exposure pathways

367   Figure 4 shows the results of the comparison between $iF_{[0-100m]}$ for bystanders and the iFs

368   predicted by USEtox for environmentally mediated general population exposure aggregating

369   inhalation and ingestion, and by dynamiCROP for general population ingestion exposure to residues

370   in harvested food crops. The differences in iFs between bystander exposure, environmentally

371   mediated population exposure and population exposure to crop residues (Figure 4) show that crop

372   residue ingestion and environmentally mediated exposure dominates total exposure from pesticides

373   applied to agricultural fields, while bystander exposure only contributes by about 0.1% to the total

374   iF (sum over all exposed populations and exposures associated with the same application scenario).

Exposure

■ Residential bystander exposure (0-100 m range)
■ Crop residue population exposure
□ Environmentally mediated population exposure
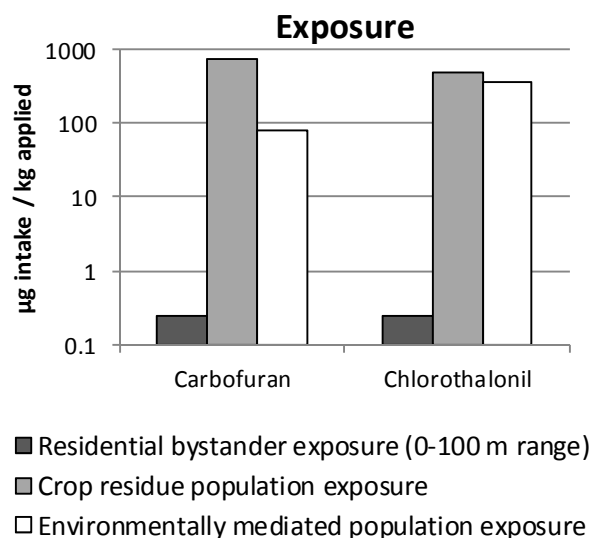
375

376 Figure 4 Intake fractions for total exposed population (all considered bystanders for bystander

377 exposure, all exposed consumers for crop residue exposure, and total population for

378 environmentally mediated exposure) per kg of pesticide applied to agricultural fields.

379

380 The question is, however, whether this kind of direct comparison between iF representing

381 different population sizes is meaningful. Indeed, bystander exposure was found to dominate

382 together with exposure via ingestion of crop residues aggregated per capita (i.e. individual) intake

383 fractions, while environmentally mediated general population exposure was found to have almost

384 no contribution to aggregated individual intake fractions (Figure 5).

385

18
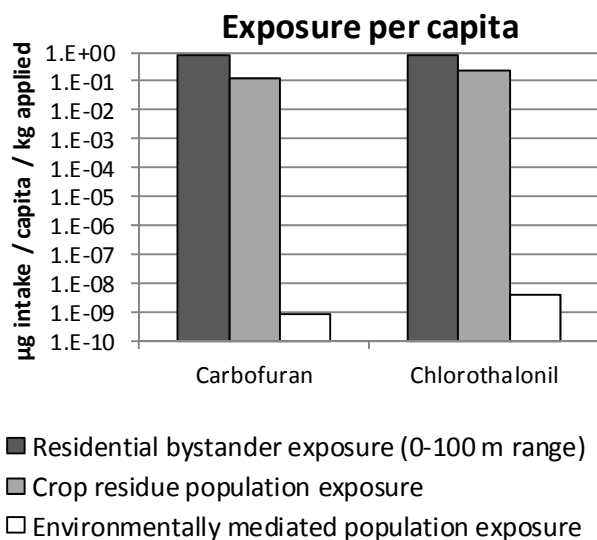
387 Figure 5 Individual (i.e. per capita) intake fractions per kg of pesticide applied to agricultural fields.

388

389 **4. Discussion**

390 **4.1. Model framework and performance**

391 The developed residential bystander model allows for predicting intake fractions for

392 different exposure ranges as a function of the distance from the field of application. Related

393 characterization factors can be estimated by multiplication of these bystander exposure results with

394 effect factors for the assessed pesticides. It is, per default, recommended for LCA practitioners to

395 apply results for the 0 to 100 m distance to the field edge range, because this range includes the full

396 residential bystander population potentially exposed to pesticide spray drift fractions and, thereby,

397 gives the most comprehensive indication of related bystander exposure. However, the fact that

398 exposure of bystanders decreases substantially after about 8 m distance from the considered field

399 edge underlines the benefit of spray buffer zones around agricultural fields which can substantially

400 reduce exposure of bystanders to spray drift (de Snoo and de Wit, 1998). Such a difference in

401 agricultural practice is of interest from an LCA perspective, particularly in the agrifood sector, and

402 our model allows this to be captured in LCA results.

403   The model was only compared to measurements for ground field crops (potatoes). The

404   model predictions were generally in good agreement with field measurements for pesticide air

405   concentrations, especially in relation to natural variability of pesticide application and spray drift

406   together with general uncertainty in measurements of spray drift. This shows that our bystander

407   exposure model provides a realistic indication for the exposure of bystanders to pesticides during

408   and after application to agricultural fields. However, further testing of the model against

409   measurements for other application such as in orchards or vineyards is required to increase the

410   validity and applicability of the model. The comparison of our bystander exposure estimates with

411   estimates of exposure from crop residues and environmentally mediated emissions shows that

412   bystander exposure does contribute to overall pesticide population intake, but that population intake

413   is generally not dominated by spray drift related exposure of bystanders, which is mainly due to the

414   small fraction of the general population that is considered to be residential bystanders living in the

415   close vicinity to agricultural fields.

416   Dividing the fate calculation part of the bystander model framework into an emission

417   fraction quantification component and an environmental fate quantification component as part of

418   the impact assessment is practical as it allows for using individual parts of the model for more

419   specific assessments, e.g. for impact assessment in modelling exposure to humans from pesticide

420   spray drift and for inventory modelling in estimating loss of pesticide from agricultural fields. As

421   for human exposure, the model currently includes inhalation-related pathways, while dermal

422   exposure from deposition of spray drift on bystanders is not included due to lack of relevant data

423   regarding deposition area, dermal uptake and related human effects information.

424   Moreover, for evaluating agricultural product systems in a life cycle perspective, the

425   inclusion of an inventory part (i.e. pesticide loss from fields after application) coupled with an

426   LCIA model that includes the fate of the spray drift outside the field is an advantage, because

427   information about specific pesticide losses from the field (e.g. spray drift, run-off, volatilization) are

428   often unknown while information about the mass of pesticide applied to the field are usually well

429   known (Rosenbaum et al., 2015). Hence, our bystander exposure model provides a consistent

430   framework linking known inventory data (mass applied) via emissions to environmental fate and

431   exposure for bystander populations. For application and use in LCA studies, iFs estimated using the

432   residential bystander model can be coupled with pesticide-specific EFs (as shown in Eq. 1) to

433   estimate pesticide-specific CFs for off-field spray drift from agricultural pesticide application. CFs

434   thereby represent potential human toxicity impacts per kg pesticide applied to the field. This aligns

435   well with recommendations that life cycle inventories should report the mass applied of each active

436   ingredient (Rosenbaum et al., 2015) in order to consistently cover all pathways relevant for

437   pesticides. Moreover, iFs for off-field spray drift can be integrated into existing LCIA toxicity

438   characterization models, but would have to be extended to cover many more existing pesticides. For

439   instance, our pathway can be included in USEtox by adapting the intake fraction matrix. Such

440   integration would then facilitate a consistent and more comprehensive assessment of human

441   exposure to toxic substances in practice.

442

4.2.   Application by LCA practitioners

444   Our bystander exposure calculations are generally based on generic input data for cases

445   where specific information on the application scenario is lacking, but can be modified to quantify

446   site- and application method-specific characterization results in cases where the LCA practitioner

447   has detailed knowledge about underlying environmental conditions and pesticide application. It was

448   found that the most influential parameters are bystander population density, wind speed, application

449   duration and individual inhalation rate. Statistics on average wind speed and application duration

450   are available or can be measured on site, while inhalation rate is traditionally well studied with

451   sound measurements of human inhalation rates, e.g. from the US Environmental Protection Agency

452   (2011). To ensure a robust quantification of bystander exposure, specific data on the population

453   density or the number of bystanders in the exposed area should be retrieved. These data will

454   essentially be specific to each area (or field).

455   The model currently includes spray drift curves for ground spraying of, e.g. potatoes,

456   cereals, and for spraying of fruit trees (see Supplementary material, Table S3) and can quantify

457   spray drift associated with these crops. Other application methods, such as aerial application and

458   hand-operated sprayers, are currently not included due to a lack of information regarding the

459   associated losses from spray drift. It is recommended for assessments of specific cases to obtain

460   spray drift curves that are as representative as possible to their case in order to get the most precise

461   results. This is, for instance, the case for assessments of specific crops and application techniques,

462   but also for assessments under different environmental and climatic conditions, which may affect

463   spray drift, e.g. as described in Kasiotis et al. (2014).

464

465   4.3.   Individual exposure compared to population exposure

466   The results showed that per capita exposure was higher for bystanders compared to per

467   capita exposure for pathways associated with environmentally mediated emissions or crop residues

468   (Figure 5), highlighting the significance of the population size considered to be exposed.

469   Essentially, this means that the sub-population qualifying as bystanders may be exposed by more

470   than twice the pesticide dose than the general population receives on average, depending on the

471   pesticide. In LCIA practice, however, this will currently not be captured since exposures are only

472   calculated on the total population level, so that we observe a "dilution" of those higher exposures as

473   shown in Figure 4. So far, toxicity LCIA models only consider exposure pathways affecting the

474   entire population (i.e. inhalation and ingestion of chemicals via food, including pesticide residues,

475   and general environmental exposure). As soon as a particular sub-population, e.g. bystanders or

476   workers, with a potentially much higher exposure than the general population is included in the

477   assessment, the impacts (or exposures) cannot simply be summed up anymore, because that will

478   render the higher impacts on the sub-population (few people with high exposure/effects) invisible

479   compared to the impacts on the general population (billions of people with very low

480   exposure/effects) and will also not capture potentially relevant differences in the dose-response for

481   low versus high exposure levels. This illustrates a known limitation to LCIA in not highlighting the

482   most exposed or vulnerable populations but instead focusing on highlighting (and comparing) total

483   population-level exposure.

484   This accentuates the need for harmonizing the assessment of different population sizes in

485   LCA as only reporting aggregated population-level intake fractions will fail to reveal the large

486   variability in the actual exposure of different populations. The lack of focus on exposure of specific

487   population groups might lead to unfavorable decision-making, because exposure exceeding

488   tolerable doses for specific populations may be overlooked if diluted in population-level exposure

489   results. This dilemma does not only occur for exposure to spray drift but for all types of exposure

490   that are predominantly targeting specific population groups (e.g. specific occupational or consumer

491   exposure settings). Hence, further research is required to allow for an adequate consideration of

492   sub-populations with higher exposures, which will also open the door to integration of occupational

493   and consumer exposure into the LCIA toxicity characterization.

494

495   **5.   Conclusions**

496   We presented a framework for assessing residential bystander exposure to pesticide

497   applications to agricultural fields which closes a gap in the exposure assessment included in LCA

498   studies where agricultural pesticides play a role. The inclusion of bystander exposure aids decision-

499   making based on LCA because previously unavailable insights about exposure of bystanders can

500   now be taken into account in LCA. The model showed a reasonable predictive capability in

501   comparison with field crops (potatoes). Our model currently provides a proof-of-concept that spray

502   drift can be included in LCIA, and the model can be used by LCA practitioners to address exposure

503   to initial pesticide spray drift. However, the model will have to be further adapted and validated

504   against data for additional crop types and other application techniques, such as aerial and hand-

505   operated spraying, should be included.

506   It was found that exposure of bystanders is limited relative to total exposure of populations

507   from ingestion of pesticide residues in crops, but that the magnitude taken in by bystanders can be

508   substantially larger than the intake of populations not living in the proximity to agricultural fields.

509   Hence, more focus on the most exposed populations as part of LCA could be beneficial to provide a

510   more comprehensive basis for decision-making where not only total population exposure, but also

511   exposure of differentiated population groups is taken into account.

512   Our bystander exposure model is available free of charge upon request from the authors.

513

Notes

515   The authors have no competing interests to declare.

516

Acknowledgements

523

Supplementary data

525   A full mathematical model description, specific details on parameters used for sensitivity

526   and uncertainty analysis, details on the estimation of indoor-outdoor pesticide concentration ratios,

527   and a list of reviewed spray drift models are provided in the Supplementary material.

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