Considering human exposure to pesticides in food products: Importance of dissipation dynamics

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ABSTRACT

The general public is continuously concerned about effects from pesticide exposure via residues in food crops. However, impacts from pesticide exposure are mostly neglected in food product-related LCAs. Time-to-harvest and dissipation from crops mainly drive residue dynamics with dissipation as the most uncertain aspect in characterization modeling. We analyzed measured half-lives (n=4513) with 95% falling between 0.6 and 29 days. With ~500 pesticides authorized alone in the EU for several hundred crops, however, experimental studies only cover few possible pesticide-crop combinations. Therefore, we estimated dissipation from measured data and provide reference half-lives for 333 pesticides applied at 20°C under field conditions. Our framework allows for detailed explorations of dietary choices in LCA with respect to health impacts from pesticide exposure via crop consumption. The next step is to include pesticide exposure via crop consumption along with improved pesticide dissipation data into existing LCIA methodologies for consideration in future LCA studies.

Keywords: Pesticides, human exposure, crop consumption, dissipation half-lives, life cycle impact assessment

1. Introduction

A main concern of the general public in various regions including Europe and the U.S. is related to long-term or chronic effects from low-level exposures to pesticides (Pretty 2005; Slovic 2010). Human population-level exposure predominantly occurs from residues in food crops, supplemented by exposure from pesticide fractions lost from agricultural fields after application (European Commission 2006; McKinlay et al. 2008). However, in almost all existing food product-related life cycle assessment (LCA) studies human and ecosystem impacts from exposure to pesticides (or other potentially toxic chemicals) are not considered at all or considered negligible (Heller et al. 2013). This is partly due to the perceived high uncertainty associated with impacts from exposure to toxic chemicals (Finnveden et al. 2009). A framework to characterize human health impacts of individual pesticides applied to different agricultural food crop types grown in Europe has been recently published (Fantke et al. 2011, 2012a). This framework is based on a set of interconnected compartments, for which pesticide fate and residues are calculated by solving s flexible set of differential mass balance equations with assumed first order kinetics by means of matrix algebra. In this study, time between pesticide application and crop harvest along with pesticide dissipation half-lives in crops have been identified as key aspects driving residue dynamics, which is consistent with similar assessments focusing on crop residue dynamics (Juraske et al. 2009; Rein et al. 2011). Whereas the influence of time to harvest has been analyzed and parameterized for existing life cycle impact assessment (LCIA) models, pesticide dissipation in food crops remains one of the most uncertain aspects in characterizing human toxicological impacts of pesticides via food crop consumption (Juraske et al. 2008; Fantke et al. 2012b). In the present study, we aim at reducing uncertainty associated with pesticide dissipation in crops, thereby also facilitating a feasible starting point for including human health impacts from pesticide exposure via food crop consumption in future LCA studies.

2. Methods

We start from following the framework proposed by Fantke et al. (2012a) to calculate human health impact scores for pesticides applied to agricultural fields in Europe. This approach is based on estimated agricultural application data for the five most extensively used pesticides per crop and country in 2003 for 24 member states of the EU as of 2004 (EU24) from a collaboration report between Eurostat and the European Crop Protection Association (European Commission 2007). Health impact scores of pesticide i applied to crop c in a specific year, $IS_{i,c}$ (DALY/year), are calculated from the total mass applied in this specific year, $m_{i,c}$ ($kg_{applied}$/year), the pesticide residue fraction in crop harvest, $hF_{i,c}$ ($kg_{residue}/kg_{applied}$), the food processing factor, PF_c ($kg_{intake}/kg_{residue}$) accounting for reduction of residues in crop harvest due to e.g. washing or cooking, the dose-response slope factor, $ß_i$ (incidence risk/kg_{intake}), and the severity factor expressed in disability-adjusted life years as a composite measure for overall population health impacts, *SF* (DALY/incidence):

$$IS_{i,c} = m_{i,c} \times hF_{i,c} \times PF_c \times \beta_i \times SF \quad \text{Eq. 1}$$

Fantke et al. (2012a) reported an overall model output uncertainty range, i.e. a variance in health impact scores, associated with contributing input variables along the impact pathway (Eq. 1) between 4.75 and 838,505 DALY/year across EU24 in 2003. Main source of uncertainty in the mass balance resulting in residual fractions of pesticides in crop harvest is the first order rate coefficient representing dissipation from crops. Hence, we focus in this study to reduce uncertainty related to dissipation in crops. Uncertainty of any model input and output variable *x* can be characterized by its squared geometric standard deviation $GSD^2(x)$ and related probability of 95% = $\{x/ GSD^2(x) < x < x \times GSD^2(x)\}$. We use $GSD^2(x_k)$ for input variables $x_k \in \{m_{i,c}, hF_{i,c}, PF_c, ß_i, SF\}$ as provided by Fantke et al. (2012a) to obtain $GSD^2(IS_{i,c})$ of model output (health impact scores $IS_{i,c}$):

$$\mathrm{GSD}^2(IS_{i,c}) = \exp\left(\sqrt{\sum_{k=1}(\ln[\mathrm{GSD}^2(x_k)])^2}\right) \quad \text{Eq. 2}$$

The uncertainty related to the calculation of pesticide residues in crop harvest is reported to be $GSD^2(hF_{i,c}) = 27$ and to correspond to a contribution of 29.6% to the output-related uncertainty of $GSD^2(IS_{i,c}) = 412.73$. To reduce $GSD^2(hF_{i,c})$ and thereby also $GSD^2(IS_{i,c})$, we systematically built an inventory database of existing experimental studies providing dissipation half-lives in agricultural food crops and other plants (Fantke and Juraske 2013). Measured dissipation half-lives from 811 scientific studies (>99% peer-reviewed) were available or calculated from concentration-time curves for 346 pesticides applied to 183 crops ($n = 4513$) with 95% of all half-lives falling between 0.6 and 29 days. However, with almost 500 pesticides authorized alone in the European Union for use on several hundred crops, analyzed experimental studies still cover only a small fraction of possible pesticide-crop combinations.

In response to this need, we developed models to estimate dissipation in crops based on the review from Fantke and Juraske (2013). We first characterized measured pesticide dissipation half-lives by describing their distribution and by determining the influence of temperature from a subset of 1030 data points with reported study condition average air temperatures. We then provided recommended geometric means of dissipation half-lives at 20°C and 95% confidence intervals for 333 reported pesticides, and used multiple imputations for substituting missing temperature data. Next, we proposed a regression-based model to predict dissipation half-lives for pesticides as a function of temperature, substance chemical class, selected substance properties and plant characteristics for all pesticides where reported data were not available. We finally evaluated model prediction performance using sums of squares of prediction residuals for excluded data. Results of these models for pesticide dissipation in crops were compared with previously used dissipation data in the assessment model framework proposed in Eq. 1 and compared in terms of model input and output uncertainty expressed in terms of GSD^2.

3. Results

Reference half-lives for 333 pesticides with reported temperatures applied at 20°C under field conditions range from 0.2 days for pyrethrins and 0.3 days for chlorsulfuron to 31 days for dalapon with 95% of all half-lives falling in the range between 1 and 18 days. Parameter estimates correct these half-lives for specific crops, temperatures, and study conditions. On average over all substances, temperature imputation only contributes with 5.1% to standard errors of half-lives. Standard errors of substance and plant parameter estimates approximately follow a decrease proportional to the inverse of the square root of the number of data points. With a minimum of 20 data points, we get a standard error of 0.08, implying a 95% confidence interval on geomean half-lives of a factor 1.5. For pesticides without reported data, we developed our final predictive model aims at estimating dissipation half-lives of pesticides as a function of their chemical class and properties. Substance class, plant species, cold storage conditions, temperature, substance molecular weight, octanol/water partition coefficient and saturation vapor pressure were taken into account as final predictor variables. All other tested substance properties (air/water and soil organic carbon/water partition coefficients, half-lives in air and soil) either did not significantly improve model accuracy or showed strong correlation with another variable. This model yields standard errors on the parameter estimates of 0.07 to 0.16 for substance classes and 0.06 to 0.15 for plant species. Implementing the new estimated dissipation half-lives into the model assessment framework proposed in Eq. 1 yields a reduced model input uncertainty in terms of a reduced $GSD^2(hF_{i,c}) = 12$, which corresponds to a reduction in variance of residues in crop harvest across pesticides and crops. Thereby, we reduced overall output-related uncertainty in terms of a reduced $GSD^2(IS_{i,c}) = 278.3$ and the contribution of residues in crop harvest (represented by $hF_{i,c}$) from initially 29.6% to now 19.5% (see Figure 1). With this, we reduced uncertainty in characterizing human exposure to pesticides via food crop consumption.

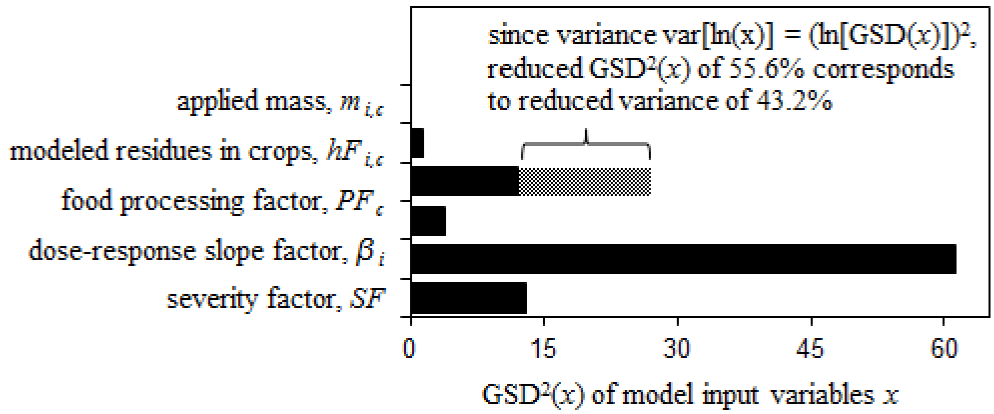

Figure 1. $GSD^2(x)$ of input variables along the pathway from pesticide application to severity of health impacts to obtain impact scores (DALY/year) due to the use of pesticides in EU24 in 2003.

Human health impact scores for use of pesticides in the EU24 countries in 2003 as applied in Fantke et al. (2012a) were re-calculated based on the newly obtained dissipation data. Overall, pesticides contributed annually to 1950 DALY/year in EU24 in 2003, to which only 13 substances applied to grapes/vines, fruit trees, and vegetables accounted for 90% of total annual health impacts. Impact scores thereby range from 0.34 DALY/year for sugar beet and 1.35 DALY/year for cereals to 724 DALY/year for grapes/vines and 1100 DALY/year for vegetables (Figure 2). The total burden per person in hours lost over lifetime was 1.5. Compared with figures for the burden from exposure to fine particulate matter in the air with 195 days or second-hand smoke in the air with 24 days (Hänninen et al. 2014), this figure is relatively low. However, uncertainties in our results – even though reduced for dissipation data in food crops – highlight the figure could be somewhat higher, ranking pesticides with other important environmental stressors in health impact terms when comparing with our upper end 95% confidence interval limit of 17.4 days burden per person over lifetime (see Figure 2).

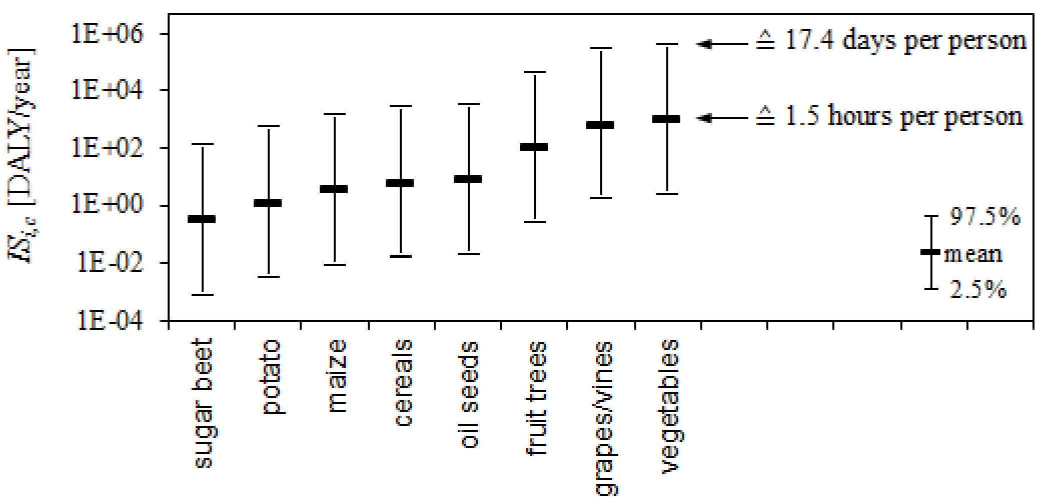

Figure 2. Mean and 95% confidence interval of impact scores (DALY/year) due to the five most extensively used pesticides on each crop class as well as sum over crops across EU24 in 2003.

4. Discussion

We developed a set of 333 comparative pesticide dissipation half-lives in food crops under reference conditions represented by an average plant, field conditions, and a temperature of 20°C (Fantke et al. 2014). These half-lives have been corrected to account for crop type, conditions, and temperature in the underlying experimental studies. This comprehensive approach, where we correct for measured temperature, reported crop species, and reported study conditions for predicting dissipation, and where we impute missing temperatures and finally study the variability of reported half-lives for each pesticide significantly reduces the uncertainty around each estimated dissipation half-life, thereby much better reflecting reference study conditions. Our additional predictive model is designed to estimate dissipation half-lives from properties of individual pesticides belonging to 14 substance classes, for which no half-lives representing reference conditions could be obtained in our study due to missing experimental data.

Considering the high variability between substances and crops, and data availability, our recommended reference half-lives along with the predictive model estimates constitute a first step towards reducing uncertainty in our assessment framework with respect to pesticide degradation in food crops. However, we acknowledge that for LCA studies, the underlying data also need to be available across pesticides and crops. Therefore, additional research is required to systematically assess the relationship between overall pesticide dissipation and degradation in crops, since typically degradation is considered as separate process in LCIA models, to further understand the influence and importance of degradation in crops as one of the main drivers of subsequent human exposure and related health impacts to be included in future LCA studies. Furthermore, reporting guidelines for measuring dissipation from food crops need to be improved with respect to providing sufficient information on environmental study conditions (e.g. reporting temperature, humidity, soil type) and residues in crops (providing enough data points to account for measurement variability and to effectively perform curve fitting for estimating dissipation kinetics). This will further increase LCIA input data quality when using experimentally-derived half-lives.

5. Conclusion

Our pesticide- and crop-specific assessment framework allows for detailed explorations of dietary choices in a LCA context with respect to human health impacts from pesticide exposure via food crop consumption. Furthermore, we reduce uncertainty related to one of the key drivers of pesticide residue dynamics in food crops in support of improving the reliability of impact assessment results. The next step is to include human exposure via crop consumption along with improved pesticide dissipation data into existing LCIA methodologies for consideration in future LCA studies. We further acknowledge that impacts from pesticides in LCAs are often dominated by impacts on biodiversity (Geiger et al. 2010) and on groundwater (Arias-Estévez et al. 2008). Hence, we emphasize the need to include pesticides into food-related LCA studies and recommend covering all relevant impact categories related to pesticides including human toxicity, ecosystem toxicity and groundwater contamination to provide an improved basis for LCA-based decision support.

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