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Life cycle toxicity assessment of pesticides used in integrated and organic production of oranges in the *Comunidad Valenciana*, Spain

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ABSTRACT

The relative impacts of 25 pesticides including acaricides, fungicides, herbicides, insecticides, and post-harvest fungicides, used in the production of oranges in Spain were assessed with current life cycle impact assessment (LCIA) tools. Chemical specific concentrations were combined with pesticide emission data and information on chemical toxicity to assess human toxicity and freshwater ecotoxicity impacts. As a case study, the relative impacts of two orange production systems in the region of Valencia, integrated pest management (IP) and organic production (OP), were assessed. The evaluation of active ingredients showed that on average acaricides have the highest human toxicity impact scores, while for freshwater ecotoxicity insecticides show the highest impact. In both impact categories the lowest impact scores were calculated for herbicides. In the production of 1 kg of orange fruits, where several kinds of pesticides are combined, results show that post-harvest fungicides can contribute more than 95% to the aggregate human toxicity impacts. More than 85% of aquatic ecotoxicity is generated by fungicides applied before harvest. The potential to reduce impacts on freshwater ecosystems is seven orders of magnitude, while impacts on human health can be reduced by two orders of magnitude. Hence, this stresses the importance of a careful pre-selection of active ingredients. In both impact categories, organic production represents the least toxic pest-control method.

1. Introduction

Spain is the largest producer of oranges in Europe and the sixth largest worldwide, with 3.4 million t of oranges harvested in 2008 (FAOSTAT, 2008). Of these, around 2.1 million t are produced on ca. 70 000 ha in the *Comunidad Valenciana* (MARM, 2008). The most harvested cultivar in this region is *navelina*, representing 41% of the total orange production in 2008. Most of the oranges produced in the region are sold for fresh consumption, while the rest (second category oranges) are used in the production of orange juice. Around 70% of all oranges allocated to fresh consumption are exported, with eastern- and central Europe as the main destinations (Sanjuán et al., 2005). A European person consumes on average 11 kg of oranges per year making this commodity the third most consumed fruit with a contribution of 14% to the total fruit diet (WHO, 2003).

As in most agricultural production systems, pesticides are also used in the cultivation of oranges in order to protect the plants from pests and consequently achieve high crop yields. Being biologically active substances with a compound-specific inherent toxicity, pesticides are considered to be significant sources of pollutants with implications concerning human health and environment integrity. Therefore, pesticide use should require permanent monitoring and evaluation (Juraske et al., 2009b). Several monitoring studies on the residues of pesticides in citrus fruits from the *Comunidad Valenciana* were carried out in the last decade and presented in the literature (Fernández et al., 2001; Berrada et al., 2006; Blasco et al., 2006; Berrada et al., 2010). Residual concentrations obtained through these studies can help in the identification of violation of threshold values (i.e. maximum residue limits) set by national or international authorities and furthermore can give an estimate on the average exposure of consumers to pesticides through a specific food commodity. Nevertheless, the evaluation of pesticide residues in plants and other environmental compartments (i.e. soil or water) by analytical methods is often limited by the high costs, the time involved, and analytical detection limits (Juraske et al., 2007a).

An alternative approach to classical laboratory analysis is life cycle impact assessment (LCIA), a method that is widely used to evaluate toxic impacts of pesticides on human- and ecosystem health (Margni et al., 2002; Antón et al., 2004; Humbert et al., 2007; Rosenbaum et al., 2008; Juraske et al., 2009b). In LCIA, the impact of toxic emissions can be assessed by fate, exposure, effect,

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and damage analysis resulting in so called characterisation factors (CF) describing the cumulative risk and potential impact per kg of emission (Jolliet et al., 2006). The total impact score of a pesticide can finally be described by the combination of the CF and the total mass release of the chemical.

The goal of this paper is to assess the relative toxicity impacts of pest-control methods on human health and freshwater aquatic ecosystems based on current LCIA tools. First, the applied methodology will be outlined in detail. As a case study, the relative impacts of two *navelina* orange (*Citrus sinensis (L.) Osbeck*) production systems: (1) integrated pest management (IP) including several pesticide treatment scenarios and post-harvest treatment, and (2) organic production (OP), were assessed. Finally optimisation options to mitigate human toxicity and environmental impacts on freshwater aquatic ecosystems are illustrated and discussed.

2. Methodology

2.1. Impact scores

The calculation of relative impact scores for pesticides on humans and the environment should include: (1) the amount of the active ingredient applied and its site of application; (2) its partitioning to the various environmental compartments; (3) its rate of degradation in each compartment; and (4) its toxicity to the species present in these compartments (Juraske et al., 2007b). The overall pesticide impact score (*IS*) of a pesticide *i* can be calculated as:

$$IS_i = CF_i \times M_i \tag{1}$$

where CF_i is the characterisation factor (impact $kg_{emitted}^{-1}$) and M_i is the mass of active ingredient *i* applied per area of land cultivated ($kg_{emitted}\ ha^{-1}$).

2.2. Human toxicity impacts

Taking disability adjusted life years (DALYs) as a measure of overall human population damage, the human toxicological characterisation factor *CF* for a pesticide *i* can be calculated according to Huijbregts et al. (2005):

$$CF_i = DF_i \times EF_i \times iF_i \tag{2}$$

where *DF* is the damage factor (DALY $case^{-1}$), *EF* is the effect factor (number of cases $kg_{emitted}^{-1}$) and *iF* is the human population intake fraction ($kg_{intake}\ kg_{emitted}^{-1}$) of a substance *i*. The latter is expressed in kg pesticide intake by food consumption per kg applied during agricultural production and was calculated according to Juraske et al. (2007a):

$$iF_i = PF \times \frac{C_{i,c} \times Y_i}{M_i} \tag{3}$$

where *PF* is the food processing factor (−), which is defined as the residue level in the processed product divided by the residue in the raw food commodity, $C_{i,c}$ is the concentration of a pesticide *i* at the time of consumption *c* ($kg\ kg^{-1}$), Y_i is the yield ($kg\ ha^{-1}$) and M_i is the mass of active ingredient applied during cultivation ($kg_{emitted}\ ha^{-1}$). Pesticide specific effect factors were obtained from Rosenbaum et al. (2008). The non-carcinogenic human health damage factor of 2.7 DALY $case^{-1}$ based on global human health statistics on life years lost and disabled was used according to Huijbregts et al. (2005).

2.2.1. Foliar application

Pesticide concentrations on and within plants treated by foliar spray application were estimated using the pesticide fate and exposure model described by Juraske et al. (2007a), which allows one to calculate the concentration of a pesticide as a function of time between application and harvest. The model takes the time between harvest and consumption, degradation of active ingredient on the plant, and loss of pesticide due to food processing like washing and peeling into account. The model, originally developed for tomatoes was modified by adjusting plant and fruit specific parameters (see Section 2.4). Degradation rates of pesticides in/on plants were used according to the methods described by Juraske et al. (2008) in which degradation rates on plant surface can be calculated from degradation rates in soil. Soil degradation data was obtained from the FOOTPRINT pesticide properties database (2010). The foliar half-life of the inorganic fungicide copper oxychloride was obtained from Ferreira et al. (2007). The time between the last foliar pesticide application and harvest is assumed to equal the pre-harvest interval (PHI) of the specific compound and the typical time between harvest and consumption was estimated to be 14 d taking into account the harvesting time, transport from the farm to the cooperative, from there to the retailers and finally to the consumer. The processing factor due to peeling of oranges ($PF = 0.08$) was obtained from Caldas et al. (2006).

2.2.2. Soil application

In the study area, all herbicides are applied with a knapsack sprayer directly onto the soil close to the root zone of the orange trees. In this case the pesticide is not deposited directly on the fruits but is potentially taken up from the soil through the roots and the stem (Juraske et al., 2009a). Pesticide concentrations in orange fruits treated by soil application were estimated using the pesticide fruit tree model presented by Paraíba (2007). This model is based on the work presented by Trapp et al. (2003) and allows for the estimation of pesticide uptake by plants from soil through the water transpiration stream and provides estimates of time-dependent pesticide concentrations in fruits. The time between the last herbicide application and harvest in the study region is four months.

2.2.3. Post-harvest treatment

Orange fruits are not only treated with pesticides during their cultivation, but also after they are harvested. This so called Post-harvest treatment (PHT) is used against storage rots caused by microbial contamination. In the study area, PHT is done by means of drench application in which the harvested fruits are drenched automatically with a fungicide solution in a chamber (drencher). Initial pesticide concentrations on orange fruits treated in a drencher were calculated on the basis of experimental data presented by King et al. (1988) providing concentrations on fruits directly after drenching (C_0) ($mg\ kg^{-1}$) which were exposed to defined application doses (*D*) ($mg\ kg^{-1}$) in the drench solution: ($C_0 = 0.0048 \times D$; $r^2 = 0.69$; $n = 6$).

2.3. Freshwater ecotoxicity impacts

For assessing the toxicological effects of pesticides on freshwater ecosystems it is necessary to imply a cause-effect chain that links emissions to impacts through environmental fate and effect (Jolliet et al., 2006). According to Rosenbaum et al. (2008), the characterisation factor for aquatic ecotoxicity (*CF*), also called ecotoxicity potential (*EP*), of a chemical *i*, providing an estimate of the potentially affected fraction of species (PAF) integrated over time and volume per unit mass of chemical emitted ($PAF\ m^3\ d\ kg^{-1}$) can be written as: