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Exposure of amateur gardeners to pesticides via the non-gloved skin per day

Martina Beránková, Jarmila Hojerová*, Linda Melegová

Faculty of Chemical and Food Technology, Slovak University of Technology, Radlinského 9, SK-812 37 Bratislava, Slovak Republic

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ABSTRACT

To predict a risk to gardeners not wearing protective gloves, the dermal absorption of three active insecticides was assessed *in vitro* using porcine ear-skin simulating 1-h handling of diluted plant protection products. Acetamiprid and Pirimicarb were found in the receptor fluid immediately after 1-h skin exposure, whereas Chlorpyrifos-methyl absorbed in the skin was not released into the receptor fluid even after 23 hours. The Estimated Gardener Exposure Level (EGEL) at 23 hours after 1-h exposure for two worst-case scenarios (i) non-gloved hands; (ii) non-gloved hands/uncovered forearms, was (i) 0.002, 0.042, and 0.057; (ii) 0.006, 0.101, and 0.135 mg/kg bw/day for Acetamiprid, Pirimicarb, and Chlorpyrifos-methyl, respectively, although the systemically available Chlorpyrifos-methyl amount, due to retention in the skin, is probably lower than determined. The Gardener Exposure Risk (GER), as a ratio of Acceptable Operator Exposure Level (databased values) to EGEL, for Acetamiprid was (i) 35 and 12-fold higher than the limit 1, so the risk via the skin is assumed to be low. Based on the GER values of (i) 0.83 and 0.18; (ii) 0.34 and 0.07 (i.e.<1) for Pirimicarb and Chlorpyrifos-methyl, respectively there is a level of concern regarding the health risk to gardeners handling pesticide products without skin protection.

1. Introduction

"Plant protection products" (PPPs), a term used by the European Food Safety Authority (EFSA, 2014), or "agricultural pesticides", a term used by the United States Environmental Protection Agency (EPA, 2017), represent the most common group of pesticide used to protect crops and desirable or useful plants in the agriculture, horticulture, forestry and viticulture as well as in home gardening. Although PPPs have been developed to function with reasonable certainty and minimal risk to human health and the environment, the published results are not always in agreement with this fact (Damalas and Eleftherohorinos, 2011). "Operators", a term used by EFSA (2014) or "handlers", a term used by EPA (2017), who carry out activities related to use of PPPs, are the persons most at risk of direct contact with these chemicals.

In recent years, a significant effort has been devoted to the quantification of exposure to PPPs for professional operators, *i.e.* farmers or contract applicators engaged in commercial crop production as a part of their job (EFSA, 2014), whereas exposure of non-professional users of PPPs is often neglected (Hojerová et al., 2015; Fevery et al., 2016). According to EFSA (2014) non-professional users are people who handle PPPs non-occupationally, for example in their home gardens. They use these products to protect plants and lawn, to grow fruits and vegetables and to control weeds that damage paths and walkways.

Most products for controlling plant pests sold for home gardening are of low actual toxicity to mammals, *i.e.* (highly) toxic or corrosive products are not authorized for non-professional use (Fevery et al., 2016). Nevertheless as they are designed to kill or control the growth and behaviour of living organisms, all PPPs pose a health risk to humans as well. When handled properly, the risk to the operator can be reduced to an acceptable level. But while professional operators are required to have a certificate of expert knowledge, amateur gardeners are usually not trained on proper use of personal protective equipments. This can result in a proportionately higher exposures of amateurs, relative to the amount of the product used (EC, 2006). Exposure of amateur gardeners to pesticides can occur due to splash or mist during many operations with PPPs. According to Harrington et al. (2005) potential exposure to non-professional PPPs is highest when diluting, mixing and loading the product into a spraying device, but applying the product and emptying/cleaning/repairing the application equipment after its use

* Corresponding author.
E-mail addresses: martina.berankova@stuba.sk (M. Beránková), jarmila.hojerova@stuba.sk (J. Hojerová).

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List of symbols

AOEL	Acceptable Operator Exposure Level
AAP	applied active pesticide
EC	European Commission
ECHA	European Chemical Agency
EFSA	European Food Safety Authority
EGEL	Estimated Gardener Exposure Level
EPA	United States Environmental Protection Agency
FTS	full-thickness skin
GER	Gardener Exposure Risk
HPLC	high-performance liquid chromatography
LoD	limit of detection
LoQ	limit of quantification
Chlorp-m	Chlorpyrifos-methyl
OECD	Org. for Economic Co-operation and Development
PPDB	Pesticide Properties Database
PPPs	plant protection products
Pow	partition coefficient in *n*-octanol/water (Log)
RF	receptor fluid
SC	*stratum corneum*
SCCS	Scientific Committee on Consumer Safety
TEC	transcutaneous electrical conductivity
UnP	unabsorbed pesticide

is also very risky. Gardeners may even be exposed without being aware of it, especially if the active pesticide enters through the skin when touching freshly treated surfaces, laundering contaminated clothing but also if pesticide-contaminated clothing is not removed promptly or not properly cleaned before being worn again. In these activities, a gardener's exposure to pesticides occurs mainly through the skin. Unlike professional operators who mostly use motorized pesticide sprayers, amateur gardeners use various pressure garden sprayers and backpack sprayers, most often holding the spray gun in their hands. Overall, hands and forearms were reported to be significantly more (47–90%) exposed to pesticides than the other body parts of the operators (Harrington et al., 2005; Rawson et al., 2005; Baldi et al., 2006; Machera et al., 2003; OECD, 2011; Fevery et al., 2016; Atabila et al., 2017).

Two surveys on the cohort of amateur gardeners were conducted in the Slovak Republic in 2015 (Hojerová et al., 2015) and 2016 (Beránková et al., 2016a) by our research group.

Paper-and-pencil questionnaires aimed at respondents' habits in handling PPPs were undertaken with customers in garden centers who confirmed that they use these products in vegetable, fruit or ornamental gardens, small vineyards and orchards or lawn care. Results of 480 completed questionnaires showed that non-professional users of PPPs are much less aware of proper hygiene practices than professional PPPs users. These findings are in line with the results of a survey on the practices of amateur gardeners in Belgium concerning the pesticides handling (Fevery et al., 2016). With regard to the protection of hands, based on the surveys' results, up to 25% of amateur gardeners completely ignore the use of personal protective gloves, 31% wear gloves while spraying PPPs and cleaning sprayers and only 44% wear gloves during all stages of PPPs handling. Moreover, the surveys revealed that most gardeners use gloves made from inappropriate material. As we have shown in our experimental study (Beránková et al., 2016b), the term “water resistant gloves” often does not mean “pesticide-resistant gloves”.

Few studies have been devoted to estimate the dermal exposure during application of non-professional PPPs (Harrington et al., 2005; Grey et al., 2006; Lessenger, 2006; Fevery et al., 2016), but no study focused on potential dermal exposure of a gardener based on dermal absorption of pesticides has been published. According to EFSA (2014) “potential dermal exposure” is the skin exposure to the chemical that would occur in the absence of clothing or personal protective equipment, while “actual dermal exposure” is the skin exposure to the chemical that would occur in the presence of clothing and/or personal protective equipment.

Thus, the first objective of this work is to predict the potential dermal exposure to active pesticides contained in commercial PPPs by the *in vitro* method under realistic conditions of use by amateur gardeners according to the results of our surveys. The second objective is to estimate the risk to the gardeners that would occur due to the systemic exposure to the active pesticide permeated through hand or hand & forearm skin in the absence of personal protective equipments.

2. Materials and methods

2.1. Pesticides and other chemicals

Three active pesticides, Acetamiprid, Pirimicarb and Chlorpyrifos-methyl, were chosen as models due to they (*i*) are insecticides (*i.e.* most often used types of PPPs) of different chemical family; (*ii*) are authorized also in the PPPs sold for the use in home gardening in the European Union (EU); (*iii*) are supplied as concentrates intended to be mixed with water and applied as a spray; (*iv*) are recommended for multiple applications during one season; and (*v*) have comparable (low) molecular weight but different hydrophilic-lipophilic properties and water solubility (see Table 1). All three active pesticides affect the nervous system by inhibition of acetylcholinesterase activity.

Acetamiprid (see Table 1) is a neonicotinoid insecticide for use as a foliar spray to control sucking-type aphids occurring on leafy vegetables, fruiting vegetables, fruit, ornamental plants and flowers. It is registered in the EU and the USA, among others. Health classification of Acetamiprid is Actual Tox. 4 (EC, 2017a). Pirimicarb (see Table 1) is a carbamate insecticide used to control aphids of various types of home garden fruit, vegetables and orchard crops. It is registered in the EU, Australia, and New Zealand, among others, but not in the USA. Health classification of Pirimicarb is Actual Tox. 3 (EC, 2017a) and Skin Sens. 1B (ECHA, 2017). Chlorpyrifos-methyl (Chlorp-m, see Table 1) is an organophosphate broad-spectrum insecticide and acaricide for the control of insects and mites on fruit and vegetables. It is registered in the EU, Australia, and USA. Health classification of Chlorp-m is Skin Sens. 1 (EC, 2017a). To monitor the skin-barrier integrity in the 3rd set of experiments (see section 2.4.3), in line with the recommendations given in the OECD Guidance Notes on dermal absorption (OECD, 2011), a reference compound caffeine (reagent-grade $\geq$98.5% from Sigma Aldrich, Steinheim, Germany), was used as an internal marker (Table 1).

Commercial formulations of PPPs include not only active substance(s), but also adjuvant chemicals to enhance their efficiency in terms of biological activity as well as to facilitate application and reaching target species, which may have an impact on dermal absorption of an active pesticide (Surgan et al., 2010; OECD, 2011; EFSA, 2014). Thus, according to EFSA (2012), ideally, dermal absorption data on PPPs should be generated on the formulated product and on concentration representative of the spray dilutions as applied to the crop. In line with this recommendation, the present study was conducted with three commercially available PPPs sold for non-professional use through the garden centers. The