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Assessment of potential dermal and inhalation exposure of workers to the insecticide imidacloprid using whole-body dosimetry in China

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ABSTRACT

In China, although improvements to the pesticide registration process have been made in last thirty years, no occupational exposure data are required to obtain a commercial license for a pesticide product. Consequently, notably little research has been conducted to establish an exposure assessment procedure in China. The present study monitored the potential dermal operator exposure from knapsack electric sprayer wheat field application of imidacloprid in Liaocheng City, Shandong Province and in Xinxiang City, Henan Province, China, using whole-body dosimetry. The potential inhalation exposure was determined using a personal air pump and XAD-2 sample tubes. The analytical method was developed and validated, including such performance parameters as limits of detection and quantification, linear range, recovery and precision. The total potential dermal and inhalation exposures were 14.20, 16.80, 15.39 and 20.78 mL/hr, respectively, for the four operators in Liaocheng and Xinxiang, corresponding to 0.02% to 0.03% of the applied volume of spray solution. In all trials, the lower part (thigh, lower leg) of the body was the most contaminated, accounting for approximately 76% to 88% of the total exposure. The inhalation exposure was less than 1% of the total exposure. Such factors as the application pattern, crop type, spray equipment, operator experience and climatic conditions have been used to explain the exposure distribution over the different parts of the body. As indicated by the calculated Margin of Exposure, the typical wheat treatment scenarios when a backpack sprayer was used are considered to be safe in terms of imidacloprid exposure.

Introduction

Pesticides are a class of products essential for sustainable agriculture and good public health. However, a general misunderstanding of the difference between the toxicity of a pesticide and the actual risk has heightened public anxiety over the use of such materials. The risk presented by a pesticide depends on its inherent toxicity and the level of exposure. Therefore, the importance of reliably assessing human exposure to pesticide risk has been growing worldwide (Palis et al., 2006). The developed countries, such as the European Union and North American countries, who have established exposure data requirements, do not allow a pesticide to be authorized unless there are specific data or adequate model predictions to show that, in normal use, the operator exposure levels would be below the acceptable operator exposure level (Hughes et al., 2006). In many developing countries, including China, no occupational exposure data

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are required to obtain a commercial license for a pesticide product, and consequently, very little research has been performed to establish exposure assessment in China.

Assessment of operator exposure to a pesticide is a very complicated task for which the difficulty fluctuates greatly depending on multiple factors, such as the application equipment (*e.g.*, hand-held or vehicle-mounted sprayer, airplane, helicopter), application rate and duration, the type of pesticide formulations (*e.g.*, powders, granules, micro-encapsulates), the climatic conditions (*e.g.*, temperature, humidity, wind speed and direction), the type of job performed by the operator, the personal protective equipment (*e.g.*, coveralls, gloves, face mask), and the training and aptitude of the applicator. All these sources of variability are grouped into "use scenarios" (Fenske and Day, 2005). The EU and USA have taken the lead in the field of occupational exposure assessment for pesticides. Under defined-use scenarios, the most important independent exposure databases, also called predictive exposure models, have been established and amended in the formal pesticide registration process. These are the Pesticide Handlers Exposure Database in the United States (PHED, 1998), the German model (Lundehn et al., 1992), the UK Predictive Operator Exposure Model (UK-POEM, 1992), the Dutch model (Van Hemmen, 1993) and the European model (Van Hemmen, 2001), which are used to estimate operator exposure.

Due to the characteristics of the small-scale farmer household economy in China, large-tonnage mechanical spraying equipment is not popular, and manual (backpack) pesticide spraying prevails in general plant protection practices in most regions of China. Moreover, the low education levels of the rural population, poor awareness of occupational hygiene, lack of information and training on pesticide safety and inadequate personal protection equipment during pesticide use place many Chinese farmers into the high pesticide exposure risk category. A large amount of attention should be paid to this situation. However, the pesticide use scenarios are different from those in developed countries, and the general exposure data cannot be readily extrapolated from the existing international predictive exposure models mentioned above. Therefore, it is imperative to carry out occupational exposure assessment for pesticides based on the fundamental realities of our own country. However, the relevant research in this field is just at the very beginning in China (Li et al., 2010; Chen et al., 2012), and research on wheat field pesticide exposure has not been reported.

The dermal route is usually the main route of exposure during the handling of pesticides, and it is considered to contribute to the greatest proportion of systemic exposure (Machera et al., 2003). In the absence of actual data on dermal absorption, most work in this field has focused on characterizing potential dermal exposure (PDE) using passive dosimetry that measures the amount of pesticide that comes into contact with the skin and clothing of workers. The measurement of PDE is a key component of risk evaluation and in helping to characterize the exposure pathway, quantifying the magnitude and extent of contamination, and evaluating the variability in sources and pesticide handler behavior (Garrido Frenich et al., 2002).

It is well known that the exposure data itself cannot be used as a risk indicator because it must be related to the acceptable exposure limits. For this, the Margin of Exposure (MOE) formula has been proposed as a useful and simple risk indicator (US EPA, 2002) that relates the acceptable exposure to the quantity absorbed by the body, which can be evaluated from values of potential exposure. The target MOE is 100 for occupational handlers. Scenarios with MOEs greater than 100 do not exceed the administrative department's level of concern.

As large agricultural provinces, the current pesticide exposure situation in Shandong and Henan provinces is representative and can generally reflect the situation in China. In the present study, field trials were conducted in farmland by four local farmers as pesticide operator volunteers. Imidacloprid, which is used for the control of many insect pests, was selected for these studies. The analytical method used for imidacloprid determination was established, and the performance parameters were fully validated. Finally, the methodology was applied to evaluate the potential dermal and inhalation exposure using whole-body dosimetry with custom-made cotton coveralls as sampling medium. The variability of the pesticide exposure and distribution pattern and evaluation of MOEs will be presented.

1. Materials and methods

1.1. Reagents and materials

Imidacloprid analytical standard (98.9% purity) was purchased from the National Pesticide Quality Supervision and Testing Center (Shenyang). The solid standard was dissolved in methanol to obtain primary stock solutions (1120 mg/L). Other working solutions were prepared by dilution with methanol. Chromatographic purity methanol was obtained from Fisher Scientific (Pittsburgh, USA), and the acetone used for extraction was analytical grade purchased from Sinopharm Chemical Reagent Beijing Co., Ltd. (Beijing, China). A commercial formulation of 15% imidacloprid soluble liquid (SL) was provided by Jiangsu Rotam Chemistry Co. Ltd. (Kunshan, China). Cotton gloves (100% cotton, 200 g/m^2) were from Beijing Kuailu Knitting Co., Ltd. (Beijing, China). An OVS tube with XAD-2 sorbent and tube holder were purchased from SKC Inc. (Eighty Four, PA, USA).

1.2. Instrumentation

All chromatographic analysis was performed on an Agilent 1200 Series HPLC (Agilent Technologies, Santa Clara, CA, USA) with an autosampler, equipped with a diode array detector and Venusil XBP C_{18} reversed-phase column (5 μm × 4.6 mm × 250 mm). For instrumentation control and data analysis, Agilent 1200 Chemstation software (Agilent Technologies, Waldbronn, Germany) was used. Standard laboratory glassware and equipment, such as a rotary evaporator (Shanghai Yarong Biochemical Instrument Factory, Shanghai, China) and overhead shaker (Shanghai Yiheng Scientific Instruments Co., Ltd., Shanghai, China), were used in the extraction procedure. The personal air sampler was AirChek 2000 from SKC Inc. (Eighty Four, PA, USA). A knapsack electric sprayer was obtained from Chaoda Instrument Co., Ltd. (CD-16B, Taizhou, China).

1.3. Field experiment

1.3.1. Application conditions

One field experiment on imidacloprid application for wheat was conducted in Liaocheng City of Shandong Province. The climatic conditions were recorded as follows: the temperature was between 21 and 22°C, relative humidity 35%–37% and average southwest wind velocity 5 km/hr. The crop was 70–80 cm high. The spray liquid was prepared by dispersing 15 g of 15% imidacloprid SL in a hydraulic knapsack electric sprayer containing 15 L of water with a single cone nozzle operating at a typical working pressure 300,000 Pa. Two experienced local farmers (operator A: female, 45 years old; operator B: male, 48 years old) performed the field experiment as volunteers. The operators started spraying, using their usual technique, with no other instructions. With the lance in front of them, approximately