



Full length article

## Transfluthrin indoor air concentration and inhalation exposure during application of electric vaporizers



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### ABSTRACT

Different household insecticide applications *via* two electric vaporizers emitting transfluthrin were realized in a full-scale experimental room under controlled air exchange rate conditions. On-line high-time resolved measurements of the gas-phase concentrations of the active substance during and immediately after the spreading periods were performed with a High Sensitivity Proton-Transfer-Reaction Mass Spectrometer (HS-PTR-MS). Experimental and modelled data from the ConsExpo 4.0 software were also compared to evaluate the sources of differences. Different application scenarios were also compared. Averaged inhaled concentrations over 1 h, 1 week, and 5 months were estimated to be 8.3, 1.8, and 1.8  $\mu\text{g}\cdot\text{m}^{-3}$ , respectively. Corresponding margins of exposures range from 1000 to 10,000, claiming for the absence of effect. Dermal and dust ingestion pathways, although roughly estimated, seems being non-negligible. This claims for a more in-depth integrated risk assessment.

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### 1. Introduction

The evaluation of indoor air contamination to environmentally-significant and health-relevant chemicals becomes a growing issue of concern, due to the current way of living that makes people spending more than 80% of their time in indoor environments (Schweizer et al., 2007). In this context, the increasing application of commercial household insecticides in indoor atmospheres is raising questions due to the potential hazardous properties of the active substances, since both exposure level and duration to those chemicals are likely to be significant.

Among the existing active substances present in household insecticides, synthetic pyrethroids belong to the insecticide family most frequently applied today (Bekarian et al., 2006; Hahn et al., 2010) due to their low toxicity for mammals, compared to organochlorine or organophosphate analogues (CDC, 2009). The use of pyrethroid insecticides is actually increasing for about ten years now (Horton et al., 2010), especially *via* the application of electric vaporizers in France (Bouvier et al., 2006). Human toxicity of pyrethroid is considered as limited (Soderlund et al., 2002) due to rapid metabolic degradation of these compounds by hydrolysis, oxidation and conjugation reactions leading to water-soluble metabolites that undergo urinary and biliary excretion (Leng et al., 1999). At levels below those inducing obvious signs of neurotoxicity (T-syndrome or CS-syndrome, Coats, 1990), several studies on animals however, show potential effects on neurodevelopment,

reproduction and immune system after the exposure to some pyrethroids (ATSDR, 2003). Pyrethroid long-term effects on human health also still remain unclear (Feo et al., 2010; Kolaczinski and Curtis, 2004) but exposure to these compounds has nevertheless been shown to cause some adverse effects, especially for children and pregnant women (ATSDR, 2003). Pyrethroid exposure thus appears to cause neurotoxicity and developmental neurotoxicity (Shafer et al., 2005), as well as adverse effects on the immune system (Rosenberg et al., 1999). Besides, many studies reveal an increased risk of cancer due to pyrethroid exposure (Ila et al., 2008; Kocaman and Topaktaş, 2009; Shukla et al., 2002). More specifically, some pyrethroids were classified by the US EPA as possible human carcinogens (US EPA (2006a,b) RED reports for permethrin and cypermethrin). Finally, these molecules are suspected to be endocrine disruptors (European Commission, 2004). Despite very frequent use of these insecticides in western countries (Bouvier et al., 2006; Grey et al., 2006), only very few studies deal with the concentration of insecticidal substances during and immediately after commercial household insecticide application (Berger-Preiss et al., 2009; Leva et al., 2009; Matoba et al., 2004; Nazimek et al., 2011; Pentamwa et al., 2011).

Consequently, this study intends to evaluate the exposure to transfluthrin during the application of two electric vaporizers in a full-scale environmental test room. Few studies (Hahn et al., 2010; Whyatt et al., 2007) demonstrated that inhalation is one of the primary routes for residential pesticide exposure. For this reason a particular attention is given to the measurements of the concentrations of the gas-phase and particulate phase in the indoor environment. An evaluation of the gas-

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phase concentration is also performed with the ConsExpo software in order to compare both experimental and modelling approach. Due to the suspected health effects of these molecules as well as the potential frequencies, levels and durations of exposure, the evaluation of the average inhaled concentrations for different durations during the application of household insecticide *via* electric vaporizers have been estimated to supplement existing long-term exposure data.

## 2. Materials and methods

### 2.1. Insecticide electric vaporizers

Two commercial electric vaporizers (5 W electric heating units) used for household treatment against mosquitoes were considered: “Raid® Electric – Fly & Mosquito Protector” and “Baygon® Genius Protector – Electric Liquid”. The active substance transfluthrin (CAS# 118712-89-3), a type I pyrethroid, is used in both commercial refills, under different formulations: solid pad refill or liquid mix refill. The active substance content (% w/w) and formulation are detailed in Table 1.

### 2.2. Test room description and conditions of application

Different application scenarios of the electric vaporizers (Table 1) were realized in the “Mechanised house for Advanced Research on Indoor Air” (MARIA) experimental house, at the Scientific and Technical Centre of Building (CSTB), Marne-la-Vallée, France (Ribéron and O’Kelly, 2002). The electric vaporizers were mounted on the supports and applied in an empty room ( $V = 32.3 \text{ m}^3$ ) of MARIA house. The ceiling is concrete painted and walls are covered with patches of painted plaster. The temperature and relative humidity were continuously measured during the experiments. Air exchange rates (AERs) were kept constant, at  $0.35 \text{ h}^{-1}$  for experiments A and C and at  $0.14 \text{ h}^{-1}$  for experiment B (Table 1).

Concerning the ventilation, the experimental air exchange rates ( $0.14$  and  $0.35 \text{ h}^{-1}$ ) correspond to realistic worst-case conditions compared to residential ventilation conditions that typically range from  $1$  to  $0.5 \text{ h}^{-1}$  for existing and new housing, respectively (Spengler et al., 2001). However, such ventilation conditions can be found in dwellings (Frederiksen et al., 2011) with defective mechanical ventilation systems (Lucas et al., 2009). More importantly, such low ventilation conditions especially occur during the night (Lucas et al., 2009) when electric vaporizers are supposed to be applied.

The vaporizers were plugged in the centre of the room at a height of about 1 m above the floor level. The application lasted 8 h according to typical night duration. The concentration of the pesticide was monitored 1 h before the beginning of the spreading period (so-called “reference situation”), during the application (increase of concentration) and once the vaporiser was unplugged, until the concentration level becomes stable and close to the initial background level (elimination phase). The vaporizers were weighted before and after their application (Table 1), in order to determine the quantity of active substance emitted, accounting for the active ingredient mass content provided by the manufacturer (Table 1). For more details about the experimental conditions the readers are referred to Vesin et al. (2013).

### 2.3. Gas-phase transfluthrin measurements

The household insecticide treatment exhibits high emission variability. Therefore, the measurements of the gaseous transfluthrin emitted by the electric vaporizer refills was performed with a High Sensitivity Proton-Transfer-Reaction Mass Spectrometer (HS-PTR-MS) (Ionicon Analytik), which provided on-line and high time-resolved measurements (Vesin et al., 2012). The HS-PTR-MS technique is based on chemical ionization of the molecules under study through  $\text{H}_3\text{O}^+$  transfer reactions, combined with subsequent mass spectrometric ion detection (Lindinger et al., 1998). The instrument is composed of an ion source in which  $\text{H}_3\text{O}^+$  are produced from pure water vapour with a hollow-cathode, a drift tube where the proton transfer reactions between  $\text{H}_3\text{O}^+$  and the molecules under study occur, and a quadrupole mass spectrometer, which differentiates the ions, according to their  $m/z$ , downstream coupled to a secondary electron multiplier detector for selective and sensitive detection.

HS-PTR-MS calibration was realized through the generation of a standard gaseous flux of transfluthrin at constant temperature under continuous controlled nitrogen flow. During the room experiments, the HS-PTR-MS was operated under the experimental conditions adjusted during the calibration step (Vesin et al., 2012).

### 2.4. Modelling of transfluthrin gas-particle partitioning

Due to the relatively low vapour pressure of transfluthrin ( $4.12 \times 10^{-4} \text{ Pa}$  at  $25 \text{ }^\circ\text{C}$ ), this Semi-Volatile Organic Compound (SVOC) is likely to be distributed between the gas-phase and the different surfaces present in the indoor environment (*i.e.*, airborne particles, settled dust, indoor surfaces). In order to evaluate the inhalation exposure following insecticide household application, the concentrations of transfluthrin in both the gas-phase and particulate phase ought to be considered. Particles of transfluthrin may actually arise due to nucleation or condensation processes that occur only if the saturated gas-phase concentration of transfluthrin ( $62 \mu\text{g}\cdot\text{m}^{-3}$  at  $25 \text{ }^\circ\text{C}$ ) is reached. A SMPS (Scanning Mobility Particle Sizer) (Grimm Technik) device scanning particles ranging from 11.1 to 1083.3 nm in diameter was used to observe eventual particle formation.

Transfluthrin is likely to be adsorbed on airborne particles (suspended matter) being already present in the room. Transfluthrin equilibrium partitioning in the air compartment between the gas-phase and airborne particles was evaluated with the model developed by Weschler and Nazaroff (2008) that has been extended in Weschler and Nazaroff (2010) and Little et al. (2012) (details are provided in supplemental material).

A modelling of the gas-phase concentration *via* the ConsExpo 4.0 software was also realized to enable a comparison with the experimental data. The model was run as a standard user would do it, only having basic information about electric vaporizer application, *viz.* the application duration (8 h), the volume of the room ( $32.3 \text{ m}^3$ ), the commercial product amount spread in the room ( $\mu\text{g}$ ), the weight fraction of active substance in the commercial product and the air exchange rate (% w/w) (Table 1). It was supposed that the pesticide is released with a constant rate during the application duration.

**Table 1**  
Characteristics of the vaporizers refills and conditions of application.

Exp.	Active substance	Commercial formulation	Commercial brand	Content (% w/w)	AER ( $\text{h}^{-1}$ )	Mass of commercial product emitted during the 8 h application (mg)
A	Transfluthrin	Solid	Raid®	13.4	0.35	18.44
B	Transfluthrin	Solid	Raid®	13.4	0.14	18.56
C	Transfluthrin	Liquid	Baygon®	0.88	0.35	463.06

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