Environment International 104 (2017) 76–82

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

Determinants of children's exposure to pyrethroid insecticides in western France

Philippe Glorennec[a,b,*], Tania Serrano[a,b], Morgane Fravallo[b], Charline Warembourg[b,c], Christine Monfort[b,c], Sylvaine Cordier[b,c], Jean-François Viel[b,d], Florent Le Gléau[e], Barbara Le Bot[a,b], Cécile Chevrier[b,c]

[a] EHESP-School of Public Health, Sorbonne Paris Cité, Rennes, France
[b] Irset INSERM-UMR1085, Research Institute for Environmental and Occupational Health, France
[c] Rennes 1 University, France
[d] Department of Epidemiology and Public Health, University Hospital, Rennes, France
[e] Labocéa, Plouzané, France

ARTICLE INFO

Keywords:
Dietary exposure
Environmental exposure
Pesticide
Biomarker
Dust

ABSTRACT

Pyrethroids are insecticides frequently used in agriculture and in the home; exposure occurs through dietary and non-dietary pathways, including indoor and outdoor environmental contamination. Our objective was to study the potential determinants of pyrethroid metabolite concentrations measured in children's urine samples and in the dust of their homes. Specifically, we measured urinary metabolites from morning spot samples of 245 six-year-old children living in Brittany (France) in 2009–2012 and from dust vacuumed from the floor of their homes. Mothers reported home insecticide use, dietary habits, sociodemographic data; residential and school proximity to agricultural crops was assessed with spatialized data. The metabolites *cis-DBCA*, *trans-DCCA*, *cis-DCCA*, *3-PBA*, and *F-PBA* were detected in 84, 95, 64, 63, and 16% of the urine samples, respectively. Permethrin, cypermethrin, cyfluthrin, deltamethrin, and tetramethrin pyrethroids were detected in 100, 56, 9, 15, and 26% of the dust samples, respectively. Multiple regression analysis suggested diet plays a role in children's exposure, in particular, the food groups “pasta, rice or semolina” (for *cis*-DCCA and F-PBA), fruit (3-PBA), “breakfast cereals and whole grain bread” (*cis*-DBCA), and the global proportion of organic food in diet (for *cis*-DBCA, *trans*-DCCA). Children with a parent occupationally exposed to pesticides were about 3-times more likely to have higher urinary concentrations of 3-PBA (OR = 2.8, 95% CI [1.2; 6.5]). Dust content was correlated mainly with household insecticide use: higher mean concentrations of permethrin (β = 0.8 [0.3; 1.3], in μg/g) and an increased risk of a detectable level of cyfluthrin (OR = 4.7 [1.7; 12.9]) were observed in home dust, for indoor use of at least twice a year. Outdoor insecticide use at least once a year was associated with detection in dust of cypermethrin (OR = 3.0 [1.3; 6.7]) and tetramethrin (OR = 3.7 [1.6; 8.3]). Three positive and one negative correlations (out of 11) between urinary metabolite concentrations and home dust contents of their possible corresponding parent compounds were observed. The strength of this study lies in its concurrent use of biomarkers, environmental measurements, and potential sources of exposure. Its limitations include the use of a single urine sample and imprecise data about pyrethroid use in local agriculture.

1. Introduction

Pyrethroids are among the most commonly used insecticides in agriculture; they are also widely used indoors in pet shampoo, lice treatment, and even insect repellent (Saillenfait et al., 2015). They are therefore frequently present both in food and in the air and dust of dwellings and thus can lead to both dietary and non-dietary exposure (Morgan, 2012). With a half-life of < 24 h, they are rapidly metabolized once absorbed to polar metabolites, eliminated primarily in urine (Leng et al., 1997). Urinary metabolite concentrations thus reflect recent exposure. The five urinary metabolites usually measured may reflect different patterns of pyrethroid exposure. The non-specific metabolite 3-phenoxybenzoic acid (3-PBA) results from exposure to cypermethrin, deltamethrin, and/or permethrin, as well as lambda-cyhalothrin, cyphenothrin, fenpropathrin, fenvalerate, fluvalinate-tau, phenothrin, and tralomethrin. The *cis*-3-(2,2-dichlorovinyl)-2,2-di-

* Corresponding author at: EHESP School of Public Health, Avenue du Pr Léon Bernard, 35043 Rennes Cédex, France.
E-mail address: philippe.glorennec@ehesp.fr (P. Glorennec).

http://dx.doi.org/10.1016/j.envint.2017.04.007
Received 21 November 2016; Accepted 17 April 2017

methylcyclopropane carboxylic acid (*cis*-DCCA) and the *trans*-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropane carboxylic acid (*trans*-DCCA) result from cyfluthrin, cypermethrin, or permethrin, and the 4-fluoro-3-phenoxybenzoic acid (F-PBA) from cyfluthrin and flumethrin. Finally, the *cis*-3-(2,2-dibromovinyl)-2,2-dimethylcyclopropane carboxylic acid (*cis*-DBCA) is a specific metabolite of deltamethrin. A recent review noted that the frequent detection of pyrethroid metabolites, especially 3-PBA and *cis*- and *trans*-DCCA, in urine samples from non-occupationally exposed populations is evidence of widespread exposure, notably of children (Saillenfait et al., 2015). In particular, 3-PBA, *cis*-DCCA, *trans*-DCCA, *cis*-DBCA, and F-PBA have been detected in urine of French adults (Frery et al., 2013) and German children (Becker et al., 2006).

Dietary determinants of the pyrethroid urinary metabolite concentrations include fruit and vegetables (Morgan, 2012), fruit juice (Morgan and Jones, 2013), and poultry (Morgan and Jones, 2013) for children, and seafood, dairy products, and cereals (Frery et al., 2013) for adults. Potential non-dietary determinants include household use—indoors and outdoors—of products containing pyrethroids as insecticides or for pets (Becker et al., 2006; Lu et al., 2006), tobacco use (Frery et al., 2013; Riederer et al., 2008), agricultural use of pyrethroids on nearby crops, and dust contamination (Becker et al., 2006).

Pyrethroid pesticides disrupt the nervous system of insects and, to a lesser degree, of mammals, and thus raise human health concerns. Although the potential risks of low levels of exposure to pyrethroids in children have not yet been adequately examined, recent studies in the general population report adverse associations between urinary concentrations of pyrethroid metabolites and neurodevelopmental outcomes among children (Oulhote and Bouchard, 2013; Viel et al., 2015).

The objective of this study was to improve our understanding of children's exposure to these insecticides by identifying the major determinants of their pyrethroid urinary metabolites, including their dietary habits and the use of products in and around the home. We also sought to assess the determinants of indoor pyrethroid levels in the home by measuring it in floor dust. We focused on urinary metabolites commonly measured and detected in Europe (Saillenfait et al., 2015), on substances that have been detected in French house dust (Blanchard et al., 2014a), and on cyfluthrin and deltamethrin because of their low volatility and common use (Saillenfait et al., 2015).These findings should help to provide guidance on developing policies to reduce and even prevent this widespread exposure among children.

2. Material and methods

2.1. Study population

The study population is a group of 245 children aged 6 years between 2009 and 2012 who participated in a neuropsychological follow-up as part of the Pélagie mother-child cohort in Brittany, France (Cartier et al., 2016; Viel et al., 2015), a region with both rural and urban areas. During this follow-up, a home visit allowed investigators to collect a first-morning-void urine sample and dust from the household vacuum cleaner. The children's parents completed a questionnaire about their dietary habits and insecticide use in and around the home. The initial population comprised 287 children, but 36 did not provide enough or any dust due to empty vacuum cleaner bags, and questionnaire data were missing for 6 more. Table 1 summarizes the population characteristics.

2.2. Selection of potential determinants of exposure

Potential determinants of pyrethroid concentrations in both urinary metabolites and dust were chosen *a priori* on the basis of a literature review. For diet, we selected the consumption of fruit and vegetables (Morgan, 2012), fruit juices (Morgan and Jones, 2013), chicken or turkey more than three times a week (Morgan and Jones, 2013), seafood, butter, cheese, yoghurt, and cereals (Frery et al., 2013). The potential non-dietary determinants selected were indoor or nearby outdoor insecticide use (Becker et al., 2006; Lu et al., 2006), exposure to environmental tobacco smoke (given the likelihood that pyrethroids are contained in tobacco leaves (Stewart and McClure, 2013) and thus present in tobacco smoke), and use of pyrethroids on local agricultural crops (provided by the Brittany Regional Agriculture Council, personal communication).

Table 1
Population characteristics and potential sources of exposure (n = 245).

Characteristic	n	%
Gender		
Female	127	52
Male	118	48
Mother's educational level[a]		
No high school diploma	32	13
Completed high school (baccalaureate)	39	16
At least 2 years postsecondary education	173	71
Body mass index ($kg \cdot m^{-2}$)		
< 14.4	61	25
14.4–15.3	57	23
15.4–16.2	67	27
> 16.2	60	24
Home: home and school near crops (< 500 m)		
Grain crops (other than corn)	216;226	88;92
Colza	74;90	30;37
Protein peas	37;57	15;-23
Fruit trees	25;32	10;13
Vegetables or flowers	37;54	15;22
Urinary cotinine ≥ 6 μg/L in child's urine sample[b]	14	6
Mother smokes[c]	54	22
One parent occupationally exposed to pesticide[d]	26	11
Use of anti-insect products at home at least once a year		
Outdoors[b]	39	16
Indoors[d]	79	33
Pesticides stored in room used in common by household	53	22
Wet floor cleaning > 2/week	70	29

Missing data: a: n = 1; b: n = 2; c: n = 3; d: n = 6.

2.3. Data collection

First-morning-void urine samples were collected during the home visit and frozen at − 20 °C in the laboratory until analysis of 3-PBA, *cis*-DCCA, *trans*-DCCA, F-PBA, and *cis*-DBCA. The details of the analytic methods have been described elsewhere (Viel et al., 2015). Briefly, 3-PBA and F-PBA metabolites in 1-mL urine samples were extracted by solid-phase extraction followed by ultra-performance liquid chromatography and triple quadrupole mass spectrometry. *Cis*-DCCA, *trans*-DCCA, and *cis*-DBCA metabolites in 2-mL urine samples were extracted, derivatized, and then analyzed by gas chromatography and then triple quadrupole spectrometry. Average recoveries were 100% ± 20%. Limits of detection (LOD) in urine were defined as the concentration with a signal to noise ratio of 3 and ranged from 0.003 μg/L for F-PBA to 0.008 μg/L for 3-PBA, whereas limits of quantification (LOQ) were defined with a signal to noise ratio of 10. The calibration curves showed good linearity with a correlation coefficient > 0.997. Regarding precision, coefficients of variations were lower or equal to 25%. For quality control, a blank (mix of pesticide-free urine) and a control sample at three concentration levels were included every 10 samples. The calibration was performed every 120 samples, and the concentration of the LOQ control level (lowest control concentration) was verified every 20 samples.

Urinary creatinine was also assessed, as was urinary cotinine (≥ 6 μg/L) (Galanti, 2008) as an indicator of environmental tobacco smoke exposure.

Household vacuum bags were collected in each home, then frozen at − 18 °C until analysis to ensure preservation of the compounds