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Trends in long term exposure to propoxur and pyrethroids in young children in the Philippines

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ABSTRACT

Background/aim: Pesticides are neurotoxic and can adversely affect children's neurobehavioral outcome. Ongoing pesticide exposure has to be monitored in the study of long term outcome of pesticide adverse effects since changes in the type and amount of exposure can influence outcome. The aim of this paper is to describe the trend in long term pesticide exposure in children through the analysis of pesticides in their hair.

Patients and methods: As part of an NIH study on the long term effects of pesticide exposure in young children, ongoing exposure to pesticides was determined by the analysis of children's hair for propoxur and pyrethroids by gas chromatography/mass spectrometry at 2, 4 and 6 years of age.

Results: There were significant changes in the prevalence and concentration of propoxur and pyrethroids in children's hair at 2, 4 and 6 years of age. At ages 2 and 4 years, the prevalence of propoxur exposure increased from 12.4% to 24.1% ($p<0.001$) but dramatically decreased to 1.7% at 6 years ($p<0.001$). For bioallethrin, the prevalence of exposure steadily increased from 2 years (0.7%, $p<0.001$) to 4 years (12.4%, $p<0.001$) and to 6 years (18.4% $p<0.001$). Exposure to transfluthrin significantly increased from 4 years (1.0%) to 6 years (9.2%, $p<0.001$).

There were also significantly higher median concentrations of bioallethrin at 2 compared to 4 years and for propoxur at 2 years compared to 6 years. Between 4 and 6 years, there was a higher median concentration of propoxur at 4 compared to 6 years and for transfluthrin and bioallethrin, at 6 compared to 4 years.

Conclusion: Changes in the prevalence and concentration of exposure to propoxur and pyrethroids in children at 2, 4 and 6 years of age are related to the progress in ambulation of young children and to changes in the formulation of home spray pesticides. Thus, periodic monitoring of pesticide exposure is necessary when studying the long term effects of pesticide exposure in the neurodevelopment of young children.

1. Introduction

There is widespread use of pesticides and vast quantities are dispersed in the environment and are subsequently found in the air, water, soil and food sources (US Environmental Protection Agency, 1999, 2010; Sekiyama et al., 2007). Human exposure to pesticides is therefore inevitable and bioaccumulation of pesticide residues in human tissues has been reported (Waliszewski et al., 2002). Pesticides are neurotoxic and have adverse effects on the children's neurobehavioral development. The child's brain in utero, at infancy and at early childhood are in a state of rapid growth and development and are therefore highly vulnerable to the toxic effects of pesticides (Eriksson, 1997; Barone et al., 2000; Bruckner 2000). House dust is a major source of pesticide exposure in young children (Clayton et al., 2003; Bradman and Whyatt, 2005). Studies have shown that more pesticides, and higher pesticide concentrations are found in household dust as compared to air, soil, and food (Lewis et al., 1999; Simcox et al., 1995). Pesticides are commonly present in house dust as a consequence of indoor spraying of pesticides (Ostrea et al., 2009; Trunnelle et al., 2013; Quirós-Alcalá et al., 2011) and proximity to agricultural areas where pesticides are used and brought indoor by drift, worker's work clothes and shoes and vehicles (Quirós-Alcalá et al., 2011; Trunnelle et al., 2013; Fenske et al., 2013; Coronado et al., 2011; Lu et al., 2000).

We are currently conducting a long term study of the pre- and postnatal effects of exposure to environmental pesticides on the neurobehavioral development of young children. At birth, we

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Table 1
Comparison of pesticide prevalence and median concentration (ng/g) for children's hair at 2, 4, and 6 years.[a]

Pesticide	Prevalence and concentration			Comparing prevalence			Comparing concentration		
	Two years (N=675)	Four years (N=711)	Six years (N=294)	Two vs four (N=649)	Two vs six (N=247)	Four vs six (N=292)	Two vs four	Two vs six	Four vs six
Propoxur	12.4 (125.8)	24.1 (29.5)	1.7 (28.6)	< 0.001	< 0.001	< 0.001	0.233	< 0.001	< 0.001
Transfluthrin	0.7 (252.3)	1.0 (107.9)	9.2 (102.4)	0.774	< 0.001	< 0.001	0.754	< 0.001	0.001
Bioallethrin	0.6 (2973.7)	12.4 (271.4)	18.4 (347.2)	< 0.001	< 0.001	0.016	< 0.001	< 0.001	0.049
Cyfluthrin	0.0	1.1 (361.3)	1.0 (376.7)	0.008[b]	0.250[b]	1.000[b]	0.012	0.109	1.000
Cypermethrin	0.0	1.3 (537.4)	1.7 (614.7)	0.004[b]	0.063[b]	1.000[b]	0.008	0.043	0.767

[a] Values given are prevalence and for positive samples, their median concentrations.
[b] Exact test.

Table 2
Agreement of pesticide prevalence and concentration (ng/g) in children's hair at 2, 4, and 6 years.[a]

Pesticide	Agreement of prevalence			Agreement of concentration		
	Two and four (N=649)	Two and six (N=247)	Four and six (N=292)	Two and four (N=649)	Two and six (N=247)	Four and six (N=292)
Propoxur	0.16 (< 0.001)	0.01 (0.857)	−0.03 (0.080)	0.15 (< 0.001)	0.02 (0.714)	−0.09 (0.092)
Transfluthrin	−0.01 (0.815)		0.04 (0.380)	−0.01 (0.815)		0.05 (0.350)
Bioallethrin	0.03 (0.027)		0.18 (0.002)	0.08 (0.032)		0.17 (0.001)
Cyfluthrin			−0.01 (0.837)			−0.01 (0.838)
Cypermethrin			0.35 (< 0.001)			0.34 (< 0.001)

When no measure is presented this is because at least one of the pesticides has no exposure for the pairs.
[a] Values given are Kappa for prevalence and Kendall's Tau for concentration and (p value).

found a high prenatal exposure rate in the newborn infants to propoxur, a carbamate and to the pyrethroids (Ostrea et al., 2009). The aim of this study is to determine the trend in long term pesticide exposure in children at 2, 4 and 6 years of age by the analysis of their hair for pesticides.

2. Materials and methods

The children are part of a cohort that was initially enrolled at birth and were born to mothers who delivered at the Bulacan Provincial Hospital in Malolos, Bulacan, an agroindustrial province of the Philippines (Ostrea et al., 2009). Their antenatal exposure to pesticides was predominantly to propoxur (a carbamate) and the pyrethroids (transfluthrin, bioallethrin, cyfluthrin and cypermethrin) as determined principally by meconium analysis (Ostrea et al., 2009). The children were followed up at 2, 4 and 6 years of age, from 2002 to 2012 and the trend in pesticide exposure was determined by the analysis of the children's hair for these pesticides. This study was approved by the Human Investigation Committees at both Wayne State University and the University of the Philippines. An informed consent was obtained from the mothers or caregivers for their participation and their children in the study.

Hair specimens from the children, about the size of a pencil eraser in diameter, were obtained from the occipital region of the head close to the scalp (Wennig, 2000; Boumba et al., 2006) and then wrapped in aluminum foil. All hair samples were packed in secondary, self-sealing polyethylene bags, labeled and kept frozen at −20 °C until the time of analysis.

3. Pesticide analysis of hair samples

The hair samples were analyzed for pesticides that were previously found at birth which included propoxur, cyfluthrin, cypermethrin, bioallethrin and transfluthrin (Ostrea et al., 2009). Unwashed hair samples were analyzed for the pesticides by gas chromatography/mass spectrometry according to previously published procedure (Bielawski et al., 2005; Posecion et al., 2006; Ostrea et al., 2006). Using spiked samples, calibration curves were linear for all pesticides with coefficients of linearity > 0.950. Optimum recovery rates ranged from 87% to 112% at a spiked concentration of 31.25 μg/g. The inter-assay and intra-assay coefficients of variation for the parent pesticides were below 11%. Limits of detection (LOD) by empirical approach (Armbruster et al., 1994) ranged from 3.60 ng/g to 488.00 ng/g hair for the various pesticides.

4. Statistical analysis

The prevalence (%) and in positive samples, the median concentrations (ng/g) of pesticides in hair at 2, 4 and 6 years of age were determined and compared by the McNemar and Wilcoxon tests, respectively. Correlation between concentrations was assessed by the Kendall's Tau test.

5. Results

There were significant changes in the prevalence and concentration of propoxur and pyrethroids in children's hair at 2, 4 and 6 years of age (Table 1). At ages 2 and 4 years, the prevalence of propoxur exposure increased from 12.4% to 24.1% ($p < 0.001$) but dramatically decreased to 1.7% at 6 years ($p < 0.001$). For bioallethrin, the prevalence of exposure steadily increased from 2 years (0.6%, $p < 0.001$) to 4 years (12.4%, $p < 0.001$) and to 6 years (18.4% ($p < 0.001$). Exposure to transfluthrin significantly increased from 4 years (1.0%) to 6 years (9.2%, $p < 0.001$). Significant increases in exposure to cyfluthrin and cypermethrin was only seen between 2 to 4 years of age and the rate of increase was small (0% to 1.1–1.7%).

There were also significantly higher median concentrations of bioallethrin at 2 compared to 4 years and for propoxur at 2 years compared to 6 years (Table 1). Between 4 and 6 years, there was a higher median concentration of propoxur at 4 compared to 6 years and for transfluthrin and bioallethrin, at 6 compared to 4 years.