Indoor Air Concentrations of Organochlorine, Organophosphate and Pyrethroid Pesticides in the US: Four Studies, Six States and Twenty Years

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Introduction

Organochlorine (OC) pesticides such as DDT, chlordane, dieldrin, heptachlor, endrin, methoxychlor, pentachlorophenol, and lindane have been widely used to control agricultural and peridomestic insect pests. By the late 1980's most agricultural and specialty market registrations for OC pesticides in the United States (US) were voluntarily withdrawn or their use severely restricted. The USEPA has estimated that chlordane and heptachlor, prior to withdrawal from the marketplace, were used to treat 24 million homes in the US[1]. Two important organophosphate (OP) insecticides, chlorpyrifos and diazinon, popularly used to control residential pests, have recently been restricted from indoor applications resulting in the pyrethroid (PYR) insecticides filling market vacancies.

Pesticides enter indoor environments by direct application or intrusion from outdoor sources. Indoors pesticides are protected from environmental degradation factors such as sunlight, moisture, and soil microbial activity and may decline more slowly than those located outdoors. As a result, concentrations may be 10 to 100 times higher indoors[2], where people spend 65-90% of their time[3]. In order to better understand exposure to pesticides, the US Environmental Protection Agency (US EPA) and others have conducted multimedia, multiresidue studies where OC, OP, and PYR pesticides were measured in homes. These studies span almost 20 years and are sufficient to observe general trends based on their descriptive statistics. The goal of this paper is to summarize the air concentrations of select residential use insecticides measured from four EPA sponsored regional studies. Differences between the airborne concentrations over time and their implications will be discussed. Ongoing research and future directions will be presented.

Materials and Methods

Several EPA sponsored studies have measured OC compounds and other pesticides in residences. The studies presented here are the Non-Occupational Exposure Study (NOPES)[4,5], the National Human Exposure Assessment Survey (NHEXAS-Maryland)[6], the Minnesota Children's Pesticide Exposure Study (MNCPES)[7], the Children's Total Exposure to Persistent Pollutants and Other Persistent Organic Pollutants (CTEPP)[8] study. Generally, these studies have collected multimedia samples (air, dust, soil, surface wipes, etc.) from homes for a spectrum of pesticide and non-pesticide compounds with an emphasis on children's exposures. Air monitoring for pesticides across all the studies utilizes modified versions of an ASTM protocol[9] developed for the NOPES. The methods employed use polyurethane foam filters (sometimes in conjunction with XAD) and pumps capable of producing flows of 3.8 L/min for 24 to 48 hours. For the purposes of this paper only airborne concentrations will be discussed. Table 1 shows for each study, the method detection limit (MDL) for select pesticide analytes. Tables 2 and 3 show the years the studies were conducted, the detection frequencies and the concentrations measured.

Results and Discussion

The detection limits varied across studies and merit consideration (Table 1). NOPES MDLs are presented as ranges based on the analysis of multimedia samples by gas chromatograph equipped with either an electron capture (non-selective) or mass spectrometric (selective) detector. The authors' indicate that lower values are conservative estimates associated with cleaner samples[3,4]. NHEXAS-MD detection limits were relatively high (»14 ng/m^3) with some mean concentrations below the reported MDL. The MNCPES and CTEPP MDLs provide the lowest estimates.

The frequency of detection (Table 2) varied between studies. The frequency of heptachlor detection in CTEPP-OH was about half of that in CTEPP-NC. Detection frequencies for both isomers of chlordane in MNCPES and CTEPP were similar to or above the NOPES as a result of lower MDLs in the later studies. Chlordane was detected from nearly all homes sampled in the CTEPP. In general, the detection frequencies for DDT, DDE, both OP compounds, and permethrin increased from 1988 to 2001, again due to increased method sensitivity. The low detection frequencies reported from the NHEXAS-MD relative to the other studies, however, are likely a result of lower sensitivity.

The airborne OC concentrations measured from homes tended to be higher in NOPES-FL than in other studies. The regional differences in heptachlor concentrations obscure any temporal trend, but the concentrations across studies are probably not different. Chlordane concentrations, which are quite high in the benchmark NOPES, were much lower in all other studies. DDT and DDE had similar concentrations across the studies. Although OP concentrations measured in NOPES-FL are very high, the mean values for the other studies were lower and probably do not differ significantly. Changes in *cis*-permethrin concentrations do not strongly suggest increased indoor airborne concentrations over time.

By as early as 1962 the use of OC compounds to control residential pests was declining due to the proliferation of resistance in cockroach populations[10]. DDT was banned in the US in 1972, more than ten years before the completion of NOPES. The stability of indoor concentrations over time suggests the translocation of DDT and DDE in the environment and intrusion into homes. Based on the relative similarity in the concentrations among the studies we might assume a similar concentration range in most homes.

The NOPES was conducted when the use of chlordane and heptachlor for termite control was quite high; intuitively, the frequency of detection and airborne concentrations for both might have been expected to be high. Findings show a dramatic increase for chlordane from a detection frequency of »70% (NOPES) to nearly 100% (CTEPP-NC) of all homes sampled. However, while more homes have measurable concentrations, the concentrations are generally from 25 to 50 times lower in the CTEPP study than those of the NOPES. These popular termiticides were voluntarily withdrawn by the manufacturer (Velsicol) in 1988. Similarly to what has been observed with DDT, chlordane and heptachlor might be redistributing to low, but ubiquitous concentrations in indoor air.

The OP compounds (primarily chlorpyrifos, diazinon and malathion) largely supplanted the OC's and were the most popular residential pesticides during the period in which all of these studies were performed. Indoor applications of chlorpyrifos and diazinon were eliminated in 2001 and 2002, respectively, as CTEPP was concluding. The detection frequencies suggest chlorpyrifos and diazinon were ubiquitous in indoor air throughout this period.

The increased use of pyrethroids in the 1990s is not reflected in the permethrin air concentrations measured in these studies. However, the mean value reported for NOPES-FL is based on a detection frequency of only 3% and may only be a function of the statistical treatment of nondetects. In addition, the relatively low vapor pressure of permethrin (1.8 X 10^{-8} mm Hg) might also result in low air concentrations. Nonetheless, there is little change between Minnesota in 1997 and Ohio in 2001.

Future studies that measure pesticide concentrations both at the national and regional level, and include extant and current use pesticides are planned. Currently, the US EPA is collaborating with HUD in the American Healthy Home Survey where 250-500 hard surface wipe samples will be analyzed for the presence of a suite of pyrethroid, OP and OC compounds. In addition, the proposed National Children's Study poses a unique opportunity to collect additional measurements. As EPA emphasis transitions from aggregate to cumulative exposures continued interest in collecting measurements for pesticides classes with differing modes of action can be expected. The continued compilation of these and other high quality studies will further clarify temporal changes in pesticide concentrations measured in air of homes and help assess the potential for human exposures.

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Table 1. Method Detection Limits (ng/m^3) for select insecticides measured from the indoor air.

	1986-1988 NOPES	1995-1996 NHEXAS	1997 MNCPES	2000-2001 CTEPP	
	City/ State				
Pesticide	**FL and MA**	**MD**	**MN**	**NC**	**OH**
Heptachlor	0.5-3.2	14.4	0.69	0.09	0.09
Chlordane	4.0-50.0	*[a]	*	0.09	0.09
a-chlordane	*	14.1	0.19	0.09	0.09
g-chlordane	*	14.8	0.41	0.09	0.09
4,4'-DDT	2.2-4.1	14.8	0.1	0.09	0.09
4,4'-DDE	1.4-3.6	15.1	0.09	0.09	0.09
Diazinon	11.0-48.0	*	0.12	0.09	0.09
Chlorpyrifos	0.5-4.5	13.9	0.1	0.09	0.09
cis-Permethrin	2.2-53.0	*	0.04	0.09	0.39

[a] * indicates chlordane isomers were combined or separated for quantification.

Table 2. The sample size (N) and detection frequency (%) of select insecticides from the indoor air of homes.

	1986-1988 NOPES[a]				1995-1996 NHEXAS		1997 MNCPES		2000-2001 CTEPP			
	FL		MA		MD		MN		NC		OH	
Pesticide	**N**	**% D[b]**	**N**	**% D**	**N**	**% D**	**N**	**% D**	**N**	**% D**	**N**	**% D**
Heptachlor	232	74	120	60	97	71	97	87	128	92	125	34
Chlordane	232	70	120	70	*[c]	*	*	*	*	*	*	*
a-chlordane	*	*	*	*	97	41	97	74	128	98	125	93
g-chlordane	*	*	*	*	97	51	97	73	128	100	125	96
4,4'-DDT	232	12	120	5	97	7	97	79	128	37	125	24
4,4'-DDE	232	5	120	17	97	5	95	73	128	34	125	35
Diazinon	232	83	120	13	*	*	73	66	128	100	125	98
Chlorpyrifos	232	95	120	30	97	80	80	93	128	100	125	98
cis-Permethrin	232	3	120	0	*	*	89	69	128	66	125	22

[a]NOPES was conducted in Jacksonville, FL and Springfield/Chicopee, ME.

[b]%D = % Detects. [c] * indicates chlordane isomers were combined or separated for quantification

Table 3. The mean concentrations (ng/m^3) of select insecticides measured from the indoor air of homes.

	1986-1988 NOPES		1995-1996 NHEXAS	1997 MNCPES	2000-2001 CTEPP	
			City/ State			
Pesticide	**FL**	**MA**	**MD**	**MN**	**NC**	**OH**
Heptachlor	130±47	17±13	36±80	1±0.8	29±67	5±28
Chlordane	263±71	117±90	*[a]	*	*	*
a-chlordane	*	*	6±12	0.3±0.2	4±9	1±3
g-chlordane	*	*	10±20	0.8±0.5	8±15	2±6
4,4'-DDT	0.8±0.5	0.5±0.1	0.8±0.5	0.3±0.3	1±8	0.2±0.2
4,4'-DDE	0.4±0.2	0.8±0.5	0.8±0.3	0.3±1	0.2±0.3	0.2±0.2
Diazinon	205±56	26±18	*	2±6	40±217	12±51
Chlorpyrifos	230±40	8±4	20±47	6±10	19±42	6±15
cis-Permethrin	1±0.7	0±0	*	0.5±2	2±5	0.5±0.8

[a] * indicates chlordane isomers were combined or separated for quantification.