Research and Development

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## **≎EPA** Project Summary

# Nonoccupational Pesticide Exposure Study (NOPES)

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The Nonoccupational Pesticide Exposure Study was the first attempt to develop a methodology for measuring the potential exposure of specified populations to common pesticides. In this study, as in other studies utilizing the Total Exposure Assessment Methodology (TEAM), the exposures were related to actual use patterns. A selected list of 32 household pesticides were evaluated in two different cities during this study.

Air samples were collected over a 24-hour period in indoor, outdoor and personal microenvironments. In addition, limited water and dermal contact samples were collected for selected homes. The study households were selected from stratified random population samples in two urbanized areas. The samples were collected over several seasons in areas contrasting a relatively high and low use of pesticides. Dietary recall, activity pattern, and pesticide use data were collected through survey questionnaires.

The report discusses the results of the study with an emphasis on the various routes of exposure (air, water, dermal, and indirectly, food) and their relative contribution to total human exposure.

This Project Summary was developed by EPA's Atmospheric Research and Exposure Assessment Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of

the same title (see Project Report ordering information at back).

#### Introduction

In 1984, Congress appropriated FY85 monies to the U.S. Environmental Protection Agency (EPA) to assess the level of pesticide exposure experienced by the general population. Occupational exposure of specific groups of pesticide users, such as farm workers and pest control operators, had been examined and characterized by previous studies. However, little was known about the general distribution of nonoccupational exposures to household pesticides. To begin to overcome this lack of knowledge, NOPES was designed to provide initial estimates of nonoccupational exposure levels and to address the nature of the variability in exposures.

NOPES was based on the Total Exposure Assessment Methodology (TEAM) approach to exposure estimation. The Agency began developing the TEAM approach in 1979 for measuring human exposure to various environmental contaminants. In a TEAM study, probability-based survey sampling procedures are combined with questionnaire data collection and modern personal monitoring techniques to obtain statistically defensible estimates of exposure levels in the general population. The initial application of this innovative approach (Wallace, 1987) was in the estimation of exposures to volatile organic compounds (VOCs).

NOPES had both methodological and analytical objectives. NOPES sought to apply the TEAM approach to a class of chemicals not previously addressed by TEAM. Therefore, the primary methodological objective of NOPES was to develop monitoring instrumentation, laboratory procedures, and survey questionnaires for a TEAM study of pesticides. The overall analytical objective of NOPES was to estimate the levels of nonoccupational exposure to selected household pesticides through air, drinking water, food, and dermal contact.

#### **Procedure**

Work on the design phase of NOPES began in 1985. Southwest Research Institute (SwRI), of San Antonio, Texas, developed the methodology for collecting air samples and analyzing them for 32 selected pesticides and pesticide degradation products. Emphasis was placed on both identifying and quantitating the target compounds. Research Triangle Institute (RTI) of Research Triangle Park, North Carolina, developed the probability-based sampling design and the questionnaires needed to collect information about pesticide use and activity patterns. The questionnaires and monitoring and analysis procedures were tested in a pilot study conducted in Jacksonville, Florida in August and September 1985.

To permit assessment of regional and seasonal variations in exposure levels, the main NOPES data collection was conducted in three phases:

- Phase I: Summer 1986 in Jacksonville, Florida.
- Phase II: Spring 1987 in Jacksonville, Florida, and Springfield and Chicopee, Massachusetts.
- Phase III: Winter 1988 in Jacksonville, Florida, and Springfield and Chicopee, Massachusetts.

The findings of EPA's National Urban Pesticide Applicator Survey and earlier studies were used to select two study areas. Jacksonville was selected as representative of an area of the country with relatively high pesticide use, and the Springfield region was selected to represent an area of low to moderate pesticide use. In both study areas, some sample members were asked to participate in all seasons of the study, whereas others were recruited only for a single season. Monitoring some people in more than one season permitted assessment of whether the overall differences observed between seasons were due to true seasonal variations or

due to random sampling variations. Short-term temporal variations were addressed by monitoring some respondents twice in the same season.

The following activities were performed for each sample member who agreed to participate in the study:

- A study questionnaire was administered.
- A personal air sampler was given to the participant to wear or keep in close proximity for 24 h.
- Two or more fixed-site air samplers were set up and run for 24 h. At least one sampler was run in the respondent's home, and at least one was run outside the home.
- At the end of the 24-h monitoring period, an activity log questionnaire was administered.

In some households, drinking water samples were collected for analyses. Dermal exposure during pesticide application events was estimated for a small number of respondents by analyzing cotton gloves worn during typical application events following the regular monitoring period.

In all phases, RTI recruited the sample households, administered the questionnaires, and statistically analyzed the questionnaire and chemical data. SwRI performed the environmental monitoring and laboratory analyses. In Phases I and II, Environmental Monitoring and Services, Inc. (EMSI), of Camarillo, California, provided overall program management and quality assurance. EPA assumed these functions in Phase III.

#### **Results and Discussion**

The second-stage (household screening) sample size was 1,501 housing units in Jacksonville and 2,472 housing units in Springfield/Chicopee. Screening information was obtained from 1,005 Jacksonville households and 1,774 Springfield households. Second-stage response rates, computed as the number of respondents divided by the number of eligible sample members, were relatively low for face-to-face household screening, ranging from 66% for the Jacksonville spring season to 84% for the Springfield/Chicopee winter season (Table 1). Second-stage nonresponse was due more to inability to contact household members during the time period allotted for screening (56% of nonresponding eligible sample

members) than to refusals (32% of nonresponding eligible sample members).

Third-stage (personal monitoring) response rates varied by study area, season, and whether sample members were single-season or multiseason subjects. Nonresponse in the third stage was primarily due to refusals to participate (73% of nonresponding eligible sample members). The two most commonly cited reasons for refusing to participate were the amount of time required and the perceived burden associated with keeping the personal sampler nearby.

The overall response rates presented in Table 1 (45% for Jacksonville and 40% for Springfield/Chicopee) are comparable to the 44% response rate experienced in the New Jersey segment of the TEAM-VOC study (Wallace, 1987). Although these response rates are low relative to those experienced in traditional areahousehold surveys, they are typical of the rates experienced in personal monitoring studies. Low personal-monitoring response rates are believed to be primarily due to the respondent burden imposed by the monitoring systems and procedures.

Tables 2 and 3 present estimated arithmetic means for indoor, outdoor, and personal air concentrations for each season in Jacksonville and Springfield/Chicopee, respectively. Figures 1 and 2 present estimated cumulative frequency distributions as log-normal probability plots for personal air exposures for two of the study pesticides, chlorpyrifos and propoxur.

Mean outdoor air concentrations were almost always lower than mean indoor and personal concentrations. Mean personal air and indoor air concentrations were usually similar. Seasonal patterns were somewhat inconsistent. However, the pesticides found at higher concentrations in Jacksonville were highest in summer, followed by spring and then winter. For Springfield/ Chicopee, the majority of the pesticides found at higher levels had higher concentrations in the spring than in the winter. For a majority of the pesticides, indoor and personal air concentrations were higher in Jacksonville than in Springfield/Chicopee, as expected. Differences between the sites were less consistent for outdoor air concentrations.

To assess the magnitude of short-term variability relative to measurement error and seasonal variations, absolute differences between pairs of indoor air

Table 1. Response Rates

		Jacks	onville		Springfield/Chicopee			
	Summer '86	Spring '87	Winter '88	Total	Spring '87	Winter '88	Total	
Second Stage Sample Size Eligible Respondents Response rate	401 363 267 74%	550 510 336 66%	550 499 402 81%	1501 1372 1005 73%	1422 1361 956 70%	1050 978 818 84%	2472 2339 1774 76%	
Third Stage First-time sample: Selected Eligible Respondents Response rate	125 120 65 54%	79 73 53 73%	95 90 55 61%	299 283 173 61%	92 89 49 55%	73 72 37 51%	165 161 86 53%	
Overall Response Rate <sup>a</sup>	40%	48%	49%	45%	39%	43%	40%	
Followup sample: Selected Eligible Respondents Response rate	  	29 29 19 66%	19 19 16 84%	48 48 35 73%	  	20 20 15 75%	20 20 15 75%	
Total: Selected Eligible Respondents	125 120 65	108 102 72	114 109 71	347 331 208	92 89 49	93 92 52	185 181 101	

aOverall response rate = (Second-stage response rate) \* (third-stage response rate) for <u>first</u> <u>time members</u> of the sample.

measurements were computed for the five most prevalent pesticides. The mean absolute differences in replicate indoor air concentrations were computed for each study area and season and compared to the mean absolute differences between duplicate indoor air readings (Table 4) The mean absolute differences between seasons in multiseason respondent indoor air concentrations were also computed and are presented in Table 4. The magnitude of the differences between estimated measurement error variability (duplicates), estimated short-term variability (replicates), and seasonal variability (multiseason respondents) varied considerably both within and between analytes. Because of the small sample size devoted to this aspect of the study and the magnitude of the variability observed, only qualitative conclusions are supported regarding the relative magnitudes of these components of variation. Measurement error variability is generally less than short-term variability, which itself is usually less than seasonal variability. Moreover, short-term and seasonal variability are generally more comparable than short-term and measurement error variability. The fact that the short-term and seasonal variations were generally comparable in

magnitude suggests that the factors contributing to short-term variations may also be major components of seasonal variations.

### Conclusions and Recommendations

Water sampling was by design only a small component of NOPES Routine sampling of public water supplies by Jacksonville and Springfield prior to NOPES had not identified any contamination by the target compounds, and water samples collected and analyzed during the NOPES pilot study also did not contain detectable levels of any analytes. Therefore, a minimal sampling effort was believed to be sufficient for estimating water exposure to the target compounds.

The small sample sizes prevent estimation of weighted population exposure estimates from these data. However, the tack of detectable levels for most analytes and the relatively low levels occasionally detected for others suggest that exposure to the NOPES target compounds from water is minimal in the two study areas.

The dermal exposure component of NOPES was primarily a pilot study of

a method for quantifying dermal exposure levels during acute exposure events. Chronic dermal exposure was not addressed. The number of events monitored was small, and events were not randomly selected, so estimated population exposure levels cannot be developed. However, analysis of the glove data does permit assessment of the method, and provides an initial impression of the relative importance of acute dermal exposure.

Dermal dose was estimated for all 16 target compound applications monitored in NOPES. It was computed by multiplying the glove concentration by the appropriate absorption factor and ranged from 0.02 µg to 16,000 µg. Daily air exposure doses were calculated as the mean personal air concentration estimates (ng/m³) from Tables 2 and 3 multiplied by 20 m³ per day of respired air. In only three of the 16 cases was the dermal dose less than the estimated daily air dose. The dermal dose was more than an order of magnitude greater than the daily air dose in more than half the cases.

Qualitative comparisons of the relative exposure contributions of air and food were possible for some of the target compounds. The relative air and food contributions were computed for daily exposures. Mean daily exposure from inhalation was estimated by multiplying the mean personal air concentration estimates (ng/m³) for each season (Tables 2 and 3) by 20 m³ air respired per day. These daily air exposure estimates were then compared to daily dietary exposure estimates. Only qualitative comparisons were supported by the data.

The NOPES air exposure data were evaluated with regard to potential chronic health effects. Both cancer and non-cancer risks were evaluated. No risks of major concern were identified.

Evaluation of NOPES results, in addition to providing important insights about the nature and magnitude of nonoccupational pesticide exposure, suggests a number of possible avenues for further research. Specific recommendations are:

 Develop guidance for conducting exposure monitoring studies and associated methodologies for assessing human non-dietary exposure to pesticides in residential settings. These follow-up studies will be designed to permit a more comprehensive analysis of the health risks associated with

Table 2. Weighted Arithmetic Mean Concentrations in Jacksonville Aira (ng/m³)

Analyte	<u>Indoor</u>			Outdoor			Personal		
	Summer	Spring	Winter	Summer	Spring	Winter	Summer	Spring	Winter
Dichlorvos	134.5	86.2	24.5	0	0	3.2	147.6	40.2	21.4
alpha-BHC	1.2	1.2	1.1	0.0	0	0.0	0.9	0.8	0.7
Hexachlorobenzene	1.3	0.4	0.3	0.2	0	0	0.9	0.4	0.4
gamma-BHC	20.2	13.4	6.0	1.3	0.5	0.6	22.1	7.0	8.5
Chlorothalonil	5.3	2.2	6.7	0.2	0.3	0.6	0.5	0.0	2.5
Heptachlor	163.4	154.9	72.2	30.2	10.7	2.8	129.1	133.7	64.2
Ronnel	0.2	0	0	0.1	0	0	0.1	0	0.0
Chlorpyrifos	366.6	205.4	120.3	16.7	3.5	2.5	280.4	182.8	118.2
Aldrin	31.3	6.8	6.9	0.2	0	0.1	19.9	38.5	6.9
Dacthal	0.2	0	0.3	0	0	0	0.6	0	0.9
Heptachlor epoxide	0.5	0.8	0.8	0.7	0.1	0	0.6	0.5	
Oxychlordane	5.2	0	6.5	0	0	0	0.0	0.5	0.1
Captan	1.9	2.2	0.1	0	0	0	0	0.1	0
Folpet	0.5	0.7	0.6	0.3	0.4	0	0.4		0.1
2,4-D esterb	1.8	0	2.5	0.0	0.4	0.8	0.7	0.4	0.8
Dieldrin	14.7	8.3	7.2	0.7	0.0	0.8	10.1	0	3.5
Methoxychlor	0.2	0.3	0.2	0	0.0	0.8	0.3	5.4	4.8
Dicofol	0	11.0	0	0	0	0.7		0.1	0.6
cis-Permethrin	0.5	1.9	1.3	0	0	0	0	0	0
trans-Permethrin	0.4	1.1	0.8	0	0		0.1	1.3	0.8
Chlordane	324.0	245.5	220.3	38.4		0	0.1	0.3	0.5
4,4'-DDT		1.0	0.5		9.5	27.3	212.0	190.7	194.8
4,4'-DDD	••	0	0.5		0	0		0.5	0.4
4,4'-DDE		0.6	0.2		0	0		0	0
ortho-Phenylphenol	96.0	70.4	59.0		0	0		0.5	0.8
Propoxur	528.5	222.3	162.5	1.2	0.0	0.1	79.7	<b>55.6</b>	39.7
Bendiocarb	85.7	5.5		10.2	0.8	2.5	315.6	141.1	142.8
Atrazine	0	5.5 0	3.4	0	0	0	51.4	4.4	3.5
Diazinon	420.7		0	0	0	0	0.3	0	0
Carbaryl		109.2	85.7	12.6	1.1	13.8	321.6	112.7	89.0
Malathion	68.1	0.4	0	0.2	0	0	28.3	0.8	0
Resmethrin	20.8	14.9	20.4	0.3	0	0.2	9.2	10.1	16.8
r Combunii	0.1	0	0	0	0	0	0.4	0	0

<sup>&</sup>lt;sup>a</sup> A weighted mean of "0" means no detectable levels were observed. A weighted mean of "0.0" means that the weighted mean was less than 0.05.

exposure to pesticides from different routes.

- 2. Conduct prospective studies to estimate pesticide concentrations in household dust in order to explore the relationship between pesticide use and exposure, and the relative importance of the dust pathway to total human exposure, especially for infants and toddlers.
- 3. Refine the dermal exposure sampling and analytical methods

required for quantifying dermal exposures and the estimation of acute and chronic pesticide exposures. These studies will attempt to estimate transfer coefficients between surface applications and the dermal and inhalation routes of exposure.

 Improve the PUF sampling technique to reduce variability in matrix spike recoveries, evaluate analytical methodology for new compounds of interest, and prepare quality assurance standards on PUF media.

5. Conduct similar NOPES studies following revision of the population survey instruments. These revisions would incorporate improvements to the original survey design, develop more appropriate stratification variables, and permit the development of a survey data base with a larger regional or national

<sup>&</sup>lt;sup>b</sup> Methyl ester in summer, butoxyethyl ester in spring and winter.

Table 3. Weighted Arithmetic Mean Concentrations in Springfield/Chicopee Aira (ng/m³)

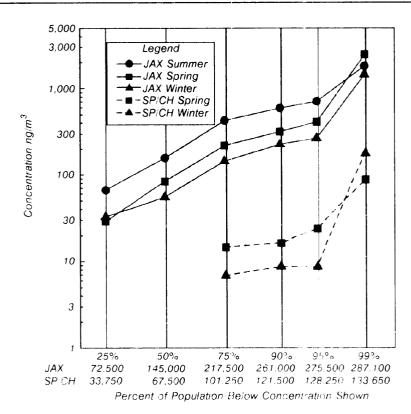
	Ind	oor	Outo	door	Personal		
Analyte	Spring	Winter	Spring	Winter	Spring	Winter	
Dichlorvos	4.3	1.5	0	0	3.7	2.1	
alpha-BHC	0.2	0	0	0	0.0	0	
Hexachlorobenzene	0	0.1	0	0	0	0.0	
gamma-BHC	0.5	9.5	0	0	0.7	5.4	
Chlorothalonil	0.1	0.1	0.4	0.8	0.8	0.1	
Heptachlor	31.3	3.6	0.3	0.1	34.7	4.6	
Ronnel	0.2	0.0	0	0	0.1	0.0	
Chlorpyrifos	9.8	5.1	13.9	0.0	7.5	5.9	
Aldrin	0	0.3	0	0	0	0.2	
Dacthal	1.6	0.3	0.9	0	2.6	0.3	
Heptachlor epoxide	0	0	0	0	0	0	
Oxychlordane	0	0	0	0	0	0	
Captan	0.1	0.0	0	0	0.1	0	
Folpet	0.7	0	0.5	0	0.7	0.0	
2,4-D butoxyethyl ester	2.1	0	0	0	0	0	
Dieldrin	1.0	4.2	0	0	0.8	0.7	
Methoxychlor	0	0	0	0	0	0	
Dicofol	0	0	0	0	7.0	0	
cis-Permethrin	0	0	0	0	0	0	
trans-Permethrin	0	0	0	0	0	0	
Chlordane	199.3	34.8	3.1	2.0	252.9	35.9	
4,4'-DDT	0.0	0.5	0	0.2	0.9	0.7	
4,4'-DDD	0	0.0	0	0	0	0	
4,4'-DDE	0.9	0.6	0	0	4.9	0.5	
ortho-Phenylphenol	44.5	22.8	1.6	0	43.4	27.3	
Propoxur	26.7	17.0	0.8	0.1	16.2	11.3	
Bendiocarb	0.2	0.4	0	0	0.3	0.2	
Atrazine	0	0	0	0	0	0	
Diazinon	48.4	2.5	8.2	9.2	10.1	1.4	
Carbaryl	0.3	0	0	0	0.1	o	
Malathion	5.0	0	0.8	0	0.5	0	
Resmethrin	0	0	0	0	0	0	

<sup>&</sup>lt;sup>a</sup> A weighted mean of "0" means no detectable levels were observed. A weighted mean of "0.0" means that the weighted mean was less than 0.05.

application. The survey instruments would incorporate more detailed activity pattern information and pesticide use applications. The data would be combined with limited monitoring data and used to validate a proposed human exposure model specifically designed to estimate exposures to several of the NOPES pesticides.

#### References

Wallace, L. A., 1987, The Total Exposure Assessment Methodology (TEAM) Study: Summary and Analysis: Volume 1. EPA/600/6-87/002. U.S. Environmental Protection Agency, Washington, DC 192 pp.



**Figure 1.** Chiorpyrifos weighted cumulative frequency distribution for personal air concentrations.

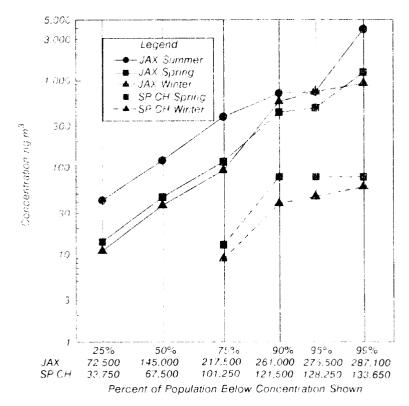


Figure 2. Propoxur weighted cumulative frequency distribution for personal air concentrations.

 Table 4. Duplicate, Replicate and Seasonal Indoor Air Concentration Differences (ng/m³)

	Duplicates			Replicates			Multiseason Respondents		
	Mean Conc.ª	Mean Abs. Diff.b	No. of Pairs	Mean Conc.ª	Mean Abs. Diff.b	No. of Pairs	Mean Conc. Over Seasons <sup>c</sup>	Mean Abs. Diff. Between Seasons <sup>d</sup>	No. of Pairs
Chlordane					,				
Jacksonville Summer Spring	55 505	2 40	6 10	271 249	98 55	8 10	369	343	19
Winter Springfield	145	60	9	129	22	9	242	114	16
Spring Winter	51 54	3 <b>8</b> 12	8 7	64 140	43 32	10 10	32	29	15
Chlorpyrifos Jacksonville									
Summer	247	38	6	362	169	8			
Spring Winter	268 187	8 17	10 9	162 152	10 <b>1</b> 19 <b>8</b>	10 9	259 122	276 114	19 16
Springfield Spring	63	16	8	34	14	10			
Winter	18	1	7	5	2	10	13	11	15
Heptachlor Jacksonville									
Summer	13	3	6	157	41	8			
Spring Winter Springfield	142 43	14 3	10 9	114 64	75 22	10 9	218 124	223 108	19 16
Spring	5	4	8	20	11	10		_	_
Winter	7	< 1	7	26	3	10	10	15	15
ortho-Phenylphenol Jacksonville									
Summer	81	29	4	91	46	5	7.5	70	47
Spring Winter	101 51	33 6	10 9	96 82	145 87	10 9	75 80	72 117	17 16
Springfield	31	U	3	02	07	J	00	,,,	,,
Spring	107	39	8	26	22	10			
Winter	54	12	7	46	23	10	34	38	15
Propoxur Jacksonville									
Summer	142	28	4	289	138	5	500	000	
Spring	378	13	10	168	137	10	529	629	17
Winter Springfield	92	10	9	51	30	9	197	184	16
Spring	48	36	8	64	18	10			
Winter	10	4	7	17	12	10	52	77	15

<sup>&</sup>lt;sup>a</sup> Unweighted mean of all matched pair data.

bUnweighted mean of the absolute differences between matched pairs.

<sup>&</sup>lt;sup>c</sup> Unweighted mean of data for two seasons from multiseason respondents. Values on the rows labelled 'Spring' are means for combined summer

and spring data; rows labelled 'Winter' are for combined spring and winter data.

d Values on rows labelled 'Spring' are the unweighted mean absciute differences between summer and spring concentrations, values on rows labelled 'Winter' are for mean absolute differences between spring and winter concentrations.

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The complete report, entitled "Nonoccupational Pesticide Exposure Study (NOPES)," (Order No. PB 90-152 224/AS; Cost: \$31 00, subject to change) will be available only from:

National Technical Information Service

5285 Port Royal Road

Springfield, VA 22161

Telephone: 703-487-4650

The EPA Project Officer can be contacted at:

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