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Chlordanes in the Indoor and Outdoor Air of Three U.S. Cities

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Indoor and outdoor concentrations of six chlordane components (*trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, *cis*-nonachlor, oxychlordane, and MC5) were measured at 157 residences, all of which were inhabited by nonsmoking individuals, in three urban areas during June 1999−May 2000. The analyses were conducted on a subset of 48 h integrated samples collected in Los Angeles County, CA, Houston, TX, and Elizabeth, NJ within the Relationship of Indoor, Outdoor, and Personal Air (RIOPA) study. Both particle-bound (PM2.5; quartz fiber filter) and vapor-phase (PUF sorbant) chlordane concentrations were separately measured by GC/EI MS after solvent extraction. The outdoor (gas + particle) total chlordane (*trans*-chlordane + *cis*-chlordane + *trans*-nonachlor + *cis*-nonachlor) concentrations ranged from 0.036 to 4.27 ng m^{-3} in Los Angeles County, from 0.008 to 11.00 ng m^{-3} in Elizabeth, and from 0.062 to 1.77 ng m^{-3} in Houston. The corresponding indoor total chlordane concentrations ranged from 0.037 to 112.0 ng m^{-3} in Los Angeles County, from 0.260 to 31.80 ng m^{-3} in Elizabeth, and from 0.410 to 38.90 ng m^{-3} in Houston study homes. Geometric mean concentrations were higher in indoor air than outdoor air (1.98 vs 0.58 ng m^{-3} in CA; 1.30 vs 0.17 ng m^{-3} in NJ; 4.18 vs 0.28 ng m^{-3} in TX), which suggests there are significant indoor sources of chlordane species in a subset of homes in each of the three cities. Calculated source strengths relate to home age, with the highest apparent indoor source strengths occurring in unattached single-family homes built during the period from 1945 to 1959. Principle indoor sources of chlordanes likely include volatilization from residues of indoor application of chlordanes and infiltration from subsurface and foundation application of chlordane-containing termiticides during home construction.

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Introduction

Exposure to chlordane species, including *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane, and MC5, is a concern due to their carcinogenic and mutagenic properties (*1*). Chlordane is listed by the World Health Organization as one of the "Dirty Dozen" Persistent Bioaccumulative Toxins (*2*). It is "possibly carcinogenic to humans" (IARC Group 2B) and/or a "probable human carcinogen, Group B2" (*3*). Under the U.S. EPA's 1996 proposed guidelines for carcinogenicity assessment, chlordane would be characterized as a "likely carcinogen" by all routes of exposure (*1*). Chlordane may also exhibit endocrine mimicry effects (*4*).

Chlordanes are components of technical chlordane, a pesticide produced and used in North America until 1997. Technical chlordane is a mixture of ∼140 compounds and contains the major components, *trans*-chlordane (TC; 13%), *cis*-chlordane (CC; 11%), *trans*-nonachlor (TN; 5%), and heptachlor (5%) as well as more than 30 less abundant chlordanes, chlordenes, and nonachlors (*5*−*9*). Throughout their history, chlordanes have been used in a number of applications including soil insecticides, termiticides, and acaricides. Chlordane was first synthesized in 1944, and after 1946 was introduced as an agricultural pesticide in the United States and around the world (*6*). It gained widespread use as a broad-spectrum pesticide from the 1940s through the 1960s. Regulations on chlordane use were initiated in 1974, and by 1983 the only remaining application of chlordane in the United States was as a termiticide, primarily in new building construction. In 1988, the termiticide registration was canceled, and sales and use in the United States were halted on April 15, 1988 (*10*). The major producer, Velsicol Chemical Co. voluntarily halted global production in 1997 (*11*). In indoor environments, chlordanes were still in active use as termiticides until the mid 1990s. Combined indoor and outdoor home use in 1990 was ∼1 million kg/year (*12*).

Due to their chemical properties, including stability, volatility, Henry's Law constant, octanol−water partition coefficient, and octanol−air partition coefficient, chlordanes are ubiquitous in the environment. They have been found in the air and biota of the Arctic and Antarctic as well as many other regions of the world (*13*−*25*). Further, these compounds tend to bioconcentrate and biomagnify in the aquatic food web (*17*, *26*−*33*). Chlordanes are also found in human serum, human breast milk, and human adipose tissue (*34*−*39*). In fact, some aboriginal peoples' dietary intakes of chlordanes (including nonachlors and oxychlordane) via consumption of traditional foods exceed the tolerable daily intake value (*40*).

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The largest sources of chlordanes to the atmosphere include volatilization from fields where chlordanes were applied, resuspension of soils containing chlordane residues (*41*), and air/water gas exchange (*42*, *43*). Sources of chlordanes could also include trans-boundary transport from regions where chlordanes may still be in active use, or volatilization of chlordanes from refuse containing chlordane residues, and/or fugitive emissions from old, unused stockpiles. Concentrations of chlordanes in outdoor air have been characterized in a number of studies (*18*, *43*−*57*). Generally, atmospheric concentrations are positively correlated with ambient air temperature indicating that air-surface exchange of chlordane species plays a major role in the observed ambient concentrations (*47*). We refer the reader to a recent overview by Bidleman et al. (*21*) of nearly 20 years of measurements of concentrations and isomer fractions measured in the Arctic atmosphere.

Concentrations of chlordanes in indoor air and their attribution to indoor and outdoor sources have been characterized in a few studies (*9*, *58*), the most comprehensive of which was conducted in 1988 (NOPES (*59*)). Comprehensive assessment of indoor and outdoor chlordane concentrations in areas with different climates and relationships of indoor to outdoor air quality is needed to quantify human exposure.

This paper presents indoor and outdoor chlordane concentrations measured in 157 homes as part of the Relationships of Indoor, Outdoor, and Personal Air (RIOPA) study. The goal of RIOPA was to gain a quantitative understanding of the impact of ambient sources of air pollutants such as VOCs, aldehydes, PM2.5, polycyclic aromatic hydrocarbons, and chlordanes on indoor air quality and human exposure by examining the concentrations of these pollutants in indoor, outdoor, and personal (breathing zone) air (*60*−*63*). RIOPA included homes in three climate zones and with a variety of house types, air exchange rates, household appliances, and activities that can influence indoor air quality. RIOPA was not a population-based study but rather a research effort which targeted homes located particularly close to, and farther from, known sources of outdoor air pollution. Chlordanes were measured in indoor and outdoor air samples collected concurrently; no personal air samples were analyzed for chlordanes. The specific objectives of this paper are (i) to assess the residential indoor and outdoor chlordane species concentrations in three geographically distinct urban areas characterized by different climate and (ii) to establish the relationship between the indoor and outdoor chlordane species concentrations.

Methods

Site Characterization and Sampling Strategy. Los Angeles County, CA, Houston, TX, and Elizabeth, NJ, were selected for the RIOPA study to represent urban areas with different climates and different types of emission sources (Weisel, 2002; Weisel et al., 2004). Los Angeles County, CA, is a metropolis interwoven by major transportation arteries and thousands of local roads that together carry a large amount of automobile traffic. Samples were collected in two different areas located northwest of the city of Los Angeles approximately 50 km apart: West Los Angeles and Santa Clarita. In West Los Angeles, the homes were located close to and farther from the intersection of the U.S. Interstates I-10 and I-405, and in Santa Clarita, homes were in the vicinity of and farther from the U.S. Interstate I-5.

Houston, TX, has a highly developed petrochemical industry. The majority of petrochemical plants and oil refineries are located northeast and east of the center of Houston, where the Houston Ship Channel provides access to the Gulf of Mexico. Samples were collected in three residential areas located in the industrial zones of Baytown, Pasadena, and Channelview.

Elizabeth, NJ, is located in central New Jersey, west of New York City. The highly developed industrial/commercial/transportation infrastructure of Elizabeth includes a marine port, numerous shipping and loading sites, an oil refinery, the Newark International Airport, and several major roads (i.e. U.S. routes 1 and 9 and U.S. Interstate I-95) with a considerable amount of diesel truck traffic. Apart from these potential emission sources, in New Jersey, as in other regions with cold winters, home heating with natural gas and oil contributes to the atmospheric particle and gas-phase pollutant concentrations during the heating season (*64*). Therefore, Elizabeth can be described as impacted by a combination of stationary and mobile sources. Sampling was carried out at homes adjacent to and farther from areas of high density of commercial traffic: Trumbull, Bayway, and along U.S. Routes 1 and 9.

Homes were selected based on their proximity to identified sources of air pollution, and in those areas participants were recruited on a volunteer basis. All participants were either English or Spanish speaking. Only nonsmokers' homes were considered for sampling. Integrated indoor and outdoor residential air samples of 48 h duration were collected simultaneously. Inside the homes, sampling equipment was placed in the main living area of the house at a height of about 1.5 m and at least 1 m from the nearest wall. The outdoor sampler was placed at ∼1.5 m height, at least 1 m from the wall of the house, and away from exhaust ducts and light and heat sources. Additional study design details are provided by Weisel et al. (*60*, *65*).

Chemical analysis of paired indoor and outdoor samples for gas-phase and particle-phase chlordane species was performed for 157 homes (61 homes in Los Angeles County, 45 homes in Houston, and 51 homes in Elizabeth), which were sampled during the period from June 1999 to May 2000. Of these 157 homes, a subset of 108 homes had a complete array of paired indoor and outdoor concentrations for both gas-phase and particle-bound chlordanes.

Air Sampling. Samples were obtained using MSP samplers (MSP Corporation, Minneapolis, MN) designed for collection of fine particulate matter, PM2.5, modified by addition of a stainless steel cylinder with an enclosed polyurethane foam plug (PUF; diameter 25 mm, height 100 mm) immediately downstream of the filter. Particle-bound chlordanes were collected on a 37 mm quartz fiber filter (QFF) downstream of a multijet 2.5 μm aerodynamic equivalent diameter cutpoint impactor. Gas-phase chlordanes were retained on the PUF. Samplers were operated at a flow rate of 10 L min^{-1}, leading to an average sample volume of 29 m^3. Air sampling face velocities averaged ∼20 cm s^{-1}. Samplers were leak checked, and flow rates and pressure drops were measured at the beginning and at the end of each sampling period. Samples were considered invalid and not included in the following analysis, if the flow changed by more than 15% during the sampling period or if the collection time was less than 42 h. Prior to sampling, PUF cartridges were hand-washed in tap water containing Alconox detergent, rinsed in deionized water followed by acetone, then sequentially extracted in Soxhlet extractors with acetone (24 h), followed by petroleum ether (24 h), and dried in a vacuum desiccator for 48 h at ambient temperature. Cleaned PUF cartridges were stored at room temperature in glass jars that had been prebaked at 400 °C and were covered with lids lined with aluminum foil. QFFs were prebaked at 550 °C for 2 h and stored at room temperature in Petri dishes lined with aluminum foil. After collecting samples, PUFs and QFFs were returned to their original containers and stored at 4 °C until analyzed.

Chemical Analysis. Prior to extraction, the PUF and QFF samples were spiked with a surrogate standard consisting of *d*10-acenaphthene, *d*10-anthracene, *d*10-fluoranthene, and *d*12-benzo[*e*]-pyrene to determine the analytical recoveries of the PAHs analyzed in a parallel portion of this project (*62*). Since no additional surrogate species were added for tracking the chlordane species, the recoveries of the deuterated PAHs were used to track the analytical recoveries of the chlordane species. The PUFs plugs were extracted statically in glass columns (i.d. 30 mm × 120 mm, with 2 mm Teflon stopcock) for 1 h with 40 mL of hot (50 °C) hexane and dichloromethane mixture (4:1 by volume). The extracts were drained into collection flasks, the PUFs were rinsed twice with 20 mL of hot hexane/DCM mixture, and the rinses were combined with the extracts. Each QFF sample was split in two portions. A 2 cm^2 portion of each filter was analyzed for organic and elemental carbon content using a thermal optical transmittance carbon analyzer (Sunset Laboratory Inc., ver. 4.0). The remaining portion of each QFF was spiked with the surrogate standard and extracted twice for 35 min with 25 mL of dichloromethane (DCM) under ultrasonic agitation.

The PUF and QFF extracts were each concentrated by rotary evaporation (Büchi RE111), followed by further concentration under a gentle nitrogen stream, and cleaned on microcolumns (i.d. 5 mm × 100 mm) of silicic acid to remove interfering polar compounds. Silica gel (60−200 mesh) was baked at 400 °C for 8 h, cooled in a desiccator for 1 h, and deactivated with 5% deionized H_2O. The column was rinsed with 2 mL of hexane/DCM (9:1 by volume). The samples were added to the column and eluted with 8 mL of 9:1 hexane/DCM. The clean extracts were reduced in volume to ∼0.1 mL by evaporation under a gentle stream of clean, dry nitrogen. An internal standard solution consisting of *d*8-naphthalene, *d*10-phenanthrene, *d*10-pyrene, and *d*12-benzo[*a*]pyrene was added to concentrated samples, and the extracts were reduced to a final volume of ∼0.05 mL (PUF) or ∼0.01 mL (QFF).

The samples were analyzed on a Hewlett-Packard 6890 gas chromatograph equipped with HP 5973 mass selective detector operated in selected ion monitoring mode. Separation of the compounds was carried out on a high-resolution capillary column (J&W Scientific; 0.25 mm i.d., 30 m long, with DB-5 as the stationary phase film, 0.25 μm film thickness). Helium (Ultrahigh Purity) was used as the carrier gas at a flow rate of 1.2 mL min^{-1}. The inlet was operated in the splitless mode at 300 °C; the injection volume was 1 μL. The temperature program was as follows: the initial temperature (50 °C) was held for 1.1 min, after which the temperature was raised using three subsequent temperature ramps (first at 25 °C min^{-1} to 125 °C, then 8 C min^{-1} to 260 °C, and finally 3.5 °C min^{-1} to 300 °C); the final temperature was held for 10 min. The analysis time for one sample was about 43 min.

Data Analysis. Data analyses, including calculations of indoor source strengths and the relative outdoor contribution to indoor concentrations, were performed using SAS statistical analysis software (version 8.0; SAS Institute, Cary, NC) running on a personal computer. The Σ-chlordane concentrations were calculated as the sum of the concentrations of *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, and *cis*-nonachlor (TC + CC + TN + CN, respectively). The *trans*-chlordane to *cis*-chlordane ratio (TC/CC = R) was also calculated and is related to the fraction of *trans*-chlordane as F_{TC} = TC/(TC + CC) = (R)/(1 + R) (*21*). Finally, indoor source strengths and relative contributions of outdoor air to the observed indoor concentrations for each compound were calculated as described later.

Quality Control. All samples including field blanks were coded with five-digit numbers, such that no information about sampling date, location of homes, or category (indoor/outdoor/field blank) was known until the results of analyses were validated. Field blanks (n = 75 and 68, PUF and QFF, respectively) were collected to determine if contamination occurred during sampling, transport, and storage; clean PUFs and QFFs were brought to the homes and placed unopened next to the samplers (indoors or outdoors) for the duration of sampling, after which they were returned to the laboratory and treated as regular samples. Σ-Chlordane mass (sum of four species: TC, CC, TN, and CN) measured in the PUF field blanks averaged 0.073 ± 0.133 ng and ranged from 0.002 to 0.836 ng, whereas in the PUF samples it averaged 112.0 ± 309.0 and ranged from 0.021 to 3120.0 ng. In the QFF field blanks, chlordane mass averaged 0.067 ± 0.151 and ranged from 0.0005 to 0.835 ng, while in the QFF samples it averaged 2.55 ± 9.37 ng and ranged from 0.001 to 117.0 ng. The method detection limits (MDL) for individual chlordane species were defined as 3 times the standard deviation of the mean compound mass in the field blanks (*61*). Because the mean chlordane species masses in the field blanks collected in different cities were similar and because no significant difference was found between the field blanks collected indoors and outdoors, media-specific MDLs were calculated using all field blanks and were applied to samples collected in each of the three cities. The MDL values for individual chlordane species are shown in Table 1.

TABLE 1. Method Limits of Detection: MDL = 3 × σ of Field Blanks (ng)

MDL: ng		gas phase	particle bound
oxychlordane	OXY	0.140	0.146
trans-chlordane	TC	0.036	0.049
cis-chlordane	CC	0.018	0.052
MC5	MC5	0.026	0.033
trans-nonachlor	TN	0.029	0.042
cis-nonachlor	CN	0.037	0.032

% greater than MDL		gas phase (%)	particle bound (%)
oxychlordane	OXY	78	6
trans-chlordane	TC	99	89
cis-chlordane	CC	99	81
MC5	MC5	99	71
trans-nonachlor	TN	99	73
cis-nonachlor	CN	94	62

Laboratory blanks (clean PUFs and QFFs) and reference standards were extracted and analyzed with every 14 samples. The mass of chlordanes in the laboratory blanks (n = 32 and 30, PUF and QFF, respectively) ranged from <MDL to 0.242 ng for the PUFs and from <MDL to 0.210 ng for the QFFs and accounted for less than 5% of the corresponding chlordane mass in the PUF and QFF samples. NIST standard reference material 1649a (Urban Dust − Organics (*66*)) was analyzed to validate the QFF analysis, and a solution of the six individual chlordane species (Supelco and Ultra Scientific) in hexane was used to validate the PUF analysis. Each material (solution of chlordanes n = 23 or NIST standard reference material n = 22) was added to the clean matrices (PUF or QFF, respectively) and then processed as a regular sample to determine the analytical accuracy of the method. Method detection limits and the percentages of samples above these limits for individual chlordane species in each phase are shown in Table 1.

In the GC/MS analysis, response factors for individual chlordane species were determined relative to the corresponding per-deuterated PAH internal standard. These were determined from the analysis of the calibration solution containing the six individual chlordane species and the deuterated PAHs which were used as internal standards. Further information about the chlordane species measured

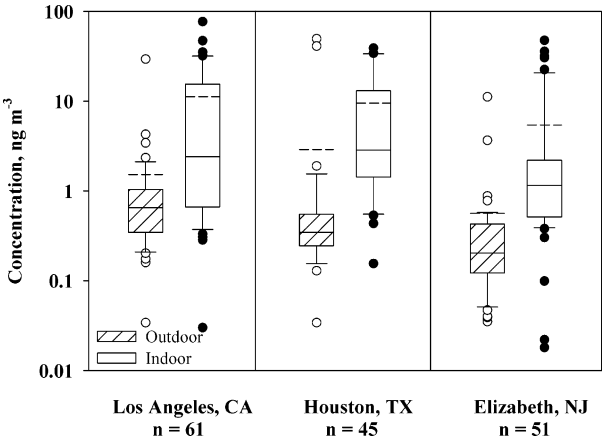

FIGURE 1. Total (gas + particle bound) sum chlordane concentrations (TC + CC + TN + CN) at paired indoor and outdoor locations. Boxes show 25th to 75th percentiles, whisker cups are 10th and 90th percentiles, and circles indicate individual values outside the 10th and 90th percentiles. Solid and dashed lines inside the boxes show the median and mean values, respectively. Outdoor concentrations are shown in hatched box and indoor concentrations in white.

in this study is listed in Table S-1 of the Supporting Information. Analytical recoveries of the PAH surrogates in the PUF ($n = 130$) and QFF ($n = 137$) samples, respectively, were 83 ± 17% and 62 ± 12% for *d*10-acenaphthene, 91 ± 16% and 69 ± 7.8% for *d*10-anthracene, 85 ± 10% and 83 ± 7.3% for *d*10-fluoranthene, and 82 ± 12% and 90 ± 8.9% for *d*12-benzo[*e*]pyrene. Though surrogate recoveries were measured, concentrations reported here were not corrected for surrogate recoveries. Likewise, the reported concentrations were not blank corrected. Assessments of capture efficiency in the PUF were conducted in Houston, TX during the summer 1999 ($n = 9$; 8 outdoor and 1 indoor), when temperatures were expected to be highest outdoors, and breakthrough potentially most significant. These experiments were conducted by placing a second PUF adsorbant trap immediately downstream of the first, to capture any losses. No significant breakthrough was observed for any of the chlordane species reported here, with less than 0.5% of each of the six chlordane species being measured in the backup PUF in all nine experiments.

Results and Discussion

Occurrence of Chlordane Species in Urban Atmospheric Environments. A summary of the outdoor and indoor total Σ-chlordane concentrations (TC + CC + TN + CN) in both the gas and particle-bound phases is presented in Figure 1. The total sum (gas + particle) chlordane concentrations in the outdoor samples ranged from 0.036 to 4.270 ng m^{-3} in Los Angeles County, from 0.008 to 11.00 ng m^{-3} in Elizabeth, and from 0.062 to 1.770 ng m^{-3} in Houston. The corresponding indoor total Σ-chlordane concentrations ranged from 0.037 to 112.0 ng m^{-3} in Los Angeles County, from 0.260 to 31.80 ng m^{-3} in Elizabeth, and 0.410 to 38.90 ng m^{-3} in Houston. Geometric mean concentrations were higher in indoor air than outdoor air (1.980 vs 0.580 ng m^{-3} in CA; 1.30 vs 0.170 ng m^{-3} in NJ; 4.180 vs 0.280 ng m^{-3} in TX), in all three cities. The outdoor Σ-chlordane concentrations were not significantly different between the three cities (unpaired Students *t*-test on log-transformed concentrations, $\alpha = 0.05$).

Variations in the chlordane concentrations in the outdoor and indoor samples were driven by gas-phase compounds, which comprised, on average, ~90% of the total chlordane mass measured in the samples (Figure 2). The geometric mean concentrations (and ranges) of individual chlordane

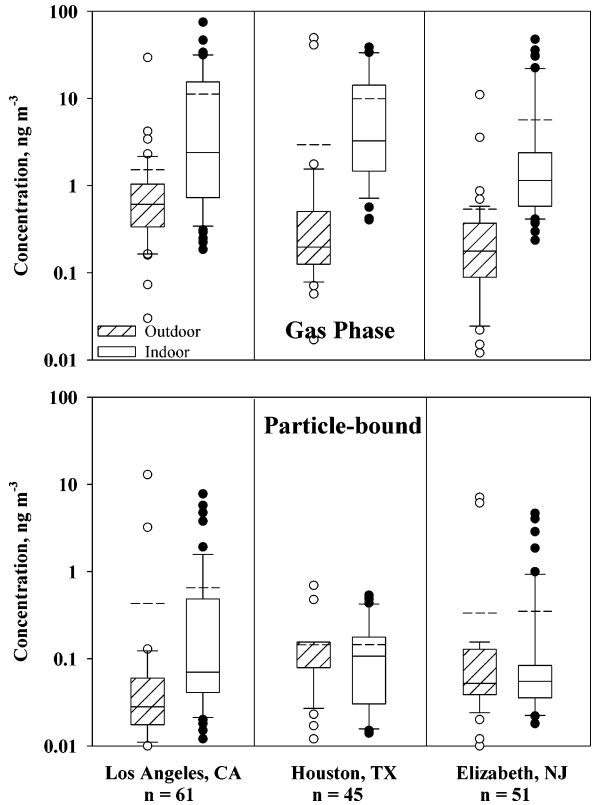

FIGURE 2. Gas- and particle-phase concentrations of sum chlordane concentrations (TC + TN + TN + CN) in paired indoor and outdoor samples at Elizabeth, NJ, Los Angeles County, CA, and Houston, TX. Boxes show 25th to 75th percentiles, whisker cups are 10th and 90th percentiles, and circles indicate individual values outside the 10th and 90th percentiles. Solid and dashed lines inside the boxes show the median and mean values, respectively. Outdoor concentrations are shown in hatched box and indoor concentrations in white.

TABLE 2. Geometric Means, Minima, and Maxima Concentrations (ng m^{-3}) of Individual Chlordane Species in Gas and Particle-Bound Phases in Indoor and Outdoor Atmospheres of Elizabeth, NJ, Los Angeles County, CA, and Houston, TX

	outdoor			indoor		
	min	geomean	max	min	geomean	max
			Particle Bound			
OXY	0.005	0.010	0.026	0.007	0.045	1.303
TC	0.002	0.006	0.027	0.003	0.024	2.131
CC	0.002	0.005	0.044	0.002	0.013	0.884
MC5	0.001	0.003	0.024	0.002	0.009	0.648
TN	0.001	0.003	0.024	0.001	0.005	0.384
CN	0.001	0.002	0.016	0.001	0.007	0.372
			Gas Phase			
OXY	0.005	0.012	0.081	0.005	0.018	0.167
TC	0.003	0.116	5.506	0.041	1.048	67.89
CC	0.003	0.082	4.301	0.005	0.653	23.93
MC5	0.001	0.021	0.398	0.002	0.118	4.216
TN	0.003	0.055	1.203	0.011	0.385	14.58
CN	0.001	0.008	0.129	0.001	0.034	1.019

species in outdoor and indoor samples are shown in Table 2.

The outdoor concentrations measured in these three cities are only slightly lower than the high levels measured in the southern United States (South Carolina in 1994−1996 and/or Alabama 1996 and 1997 (*53, 9*)) and yet higher than the levels measured over and near the Laurentian Great Lakes

TABLE 3. ΣChlordane Concentrations (ng m^{-3}) and *trans*-/*cis*-Chlordane Ratios in Outdoor Air: A Latitudinal Gradient across North America

site	year of sampling	Σ-chlordane (ng m^{-3})	F_{TC}	reference
Alert, Canada	1997	0.0015	0.38	*57*
Eagle Harbor, MI	1995−1997	0.011[a]	0.52	*55, 9*
Sturgeon Pt., NY	1995−1997	0.028[a]	0.48	*55, 9*
Elizabeth, NJ	1999−2000	0.271[b]	0.57	this work
Jersey City, NJ	1998−1999	0.074[b]	0.56	*75*
Sandy Hook, NJ	1998−1999	0.064[b]	0.54	*75*
New Brunswick, NJ	1997−1999	0.092[b]	0.54	*75*
Columbia, SC	1994−1995	0.280[b]	0.66	*53*
Houston, TX	1999−2000	0.404[b]	0.54	this work
NW Alabama	1996−1997	0.098	0.65	*9*
Los Angeles County, CA	1999−2000	0.616[b]	0.58	this work
U.S. Cornbelt air, OH, PA, and IN	1996−1998	0.050[b]	0.47	*76*
technical chlordane			0.54	*9*
technical chlordane			0.56	*77*
predicted air in equilibrium with technical chlordane at 20 °C			0.61	*9*
predicted air at equilibrium with technical chlordane at 25 °C			0.65	*53*

[a] Sum chlordane equals *trans*-chlordane + *cis*-chlordane + *trans*-nonachlor + *cis*-nonachlor except where denoted by *a* which indicates that *cis*-nonachlor was not included. [b] Indicates geometric mean instead of arithmetic mean. [c] Where F_{TC} = TC/(TC + CC) = (R)/(1 + R) and TC/CC = R.

((*43, 55*) Table 3). The outdoor concentrations reported here are an order of magnitude or two higher than measurements taken within approximately 3 years in Arctic, high Arctic, and Antarctic atmospheres (*21, 54*).

The indoor air concentrations of chlordane species measured in this portion of the RIOPA study were comparable to those measured in the Non-Occupational Pesticide Exposure Study (NOPES (*59, 67*)). NOPES measured levels of 32 pesticides in 24 h samples obtained inside and outside homes located in two cities in 1988. Approximately 70 homes in Jacksonville, FL were monitored in three seasons, and approximately 50 homes in Springfield, MA were monitored in two seasons. The indoor concentrations of chlordane ranged from 220.0 to 324.0 ng/m^3 in Jacksonville and from 34.80 to 199.0 ng/m^3 in Springfield. For both cities, average indoor chlordane concentrations in air and dust were higher than corresponding outdoor concentrations in air and soils (*59, 67*), by a factor of 10−100.

A few more recent measurements made in areas of historical agricultural (outdoor) chordane use show high indoor concentrations of these compounds. Jantunen et al. (*9*) found concentrations of individual chlordane species which averaged TC 30.0 ± 28.0, CC 18.0 ± 19.0, and TN 9.0 ± 10.0 ng m^{-3} in the indoor air of five homes in the Muscle Shoals area of Alabama. In an earlier study, Anderson and Hites (*68*) reported indoor and outdoor concentrations of chlordanes at 12 homes in Bloomington, IN from November 1985 to October 1986. The indoor concentrations for TC were much higher, ranging up to 29.0 ng m^{-3} and averaging 7.3 times greater than in outdoor air.

Wallace et al. (*70*) performed a more detailed investigation into a single home from the study of Anderson and Hites (*68, 69*). The home had elevated levels of aldrin and dieldrin, both prior to and after several remediation attempts (from September 1987 through April 1995). They reported high indoor to outdoor concentration ratios of compounds that were used indoors as pesticides, including chlordanes, along with a trend of increasing air concentrations from second floor to the basement of the house. Even after attempts to remediate the aldrin and dieldrin contamination, high indoor concentrations of TC, CC, and TN were found in the basement of this home (2.50, 3.30, and 0.30 ng m^{-3}, respectively). Indoor-to-outdoor ratios of greater than 16 for TC and 18 for CC, along with downstairs-to-upstairs ratios of 6 and 3, respectively, suggested that the primary source of chlordanes was volatilization from the foundation/basement of this home.

Relationship between the Indoor and Outdoor Chlordane Concentrations. Plots of indoor concentrations vs outdoor concentrations of individual chlordane compounds are presented in Figure 3. Points above the 1:1 line indicate homes for which the indoor concentrations are greater than outdoor concentrations. In these homes, indoor sources make a significant contribution to indoor air concentrations. Points below the 1:1 line can occur in the absence of indoor sources or in the presence of indoor sources when the species penetration through the building envelope is less than 100% or the loss rate indoors is significantly greater than zero. Such plots for Σ-chlordanes show that for 99 out of 108 homes that had paired indoor and outdoor total (gas + particle) Σ-chlordane concentrations, indoor levels were greater than outdoor levels usually more than 10 times higher inside than out and up to almost 100-fold higher in a few homes.

On an individual compound basis, indoor total (gas + particle bound) concentrations typically exceed outdoor concentrations. For example, in 103 out of 112 homes for which there were concentrations above the MDL for gas and particle phases, both indoors and out, the data for TC lays above the 1:1 line. Of these, there were 95 homes for which the indoor concentration was a factor of 2 or more greater than the corresponding outdoor concentration, 46 homes for which the indoor concentration of TC was a factor of 10 or more greater than the corresponding outdoor concentration, and 7 homes for which the indoor concentration of TC was a factor of 100 or more greater than the corresponding outdoor concentration. For one house in New Jersey sampled during winter, the indoor concentration of TC was a factor of 474 greater than the outdoor concentration. Likewise, in 100 out of 112 homes the indoor concentration of CC was greater than the outdoor concentration. Of these, there were 84 homes for which the indoor concentration was greater than 2-fold more than the outdoor concentration, 35 homes for which the indoor concentration of CC was more than 10 times greater than the corresponding outdoor concentration, and 6 homes for which the indoor concentration was more than 100-fold greater than the outdoor concentration. In the same home for which the TC indoor concentration was 474 times the outdoor concentration, the indoor concentration of CC was 489 times the outdoor concentration. In this home, the extremely high ratio for each of these compounds was driven by low outdoor concentrations in conjunction with moderately high indoor concentrations.

Different behaviors among individual compounds in the paired indoor/outdoor samples can be seen when indoor concentrations of individual compounds are plotted against corresponding outdoor concentrations. The substantial data scattering observed in each city suggests complex source/sink relationships for these compounds in the indoor air. Dispersion of the data points above the 1:1 line illustrates the importance of indoor sources. Strength of the indoor/outdoor relationships for chlordanes in each city are not explained by the air exchange rate, nor the temperature (either indoor or outdoor), nor by the difference in temperature between indoors and outdoors.

Indoor Source Strengths. Assuming perfect, instantaneous indoor mixing, indoor concentrations can be described by the following mass balance equation

$$V\mathrm{d}C_{\mathrm{in}}/\mathrm{d}t = P\alpha VC_{\mathrm{out}} - V\alpha C_{\mathrm{in}} + S - kVC_{\mathrm{in}} \qquad (1)$$

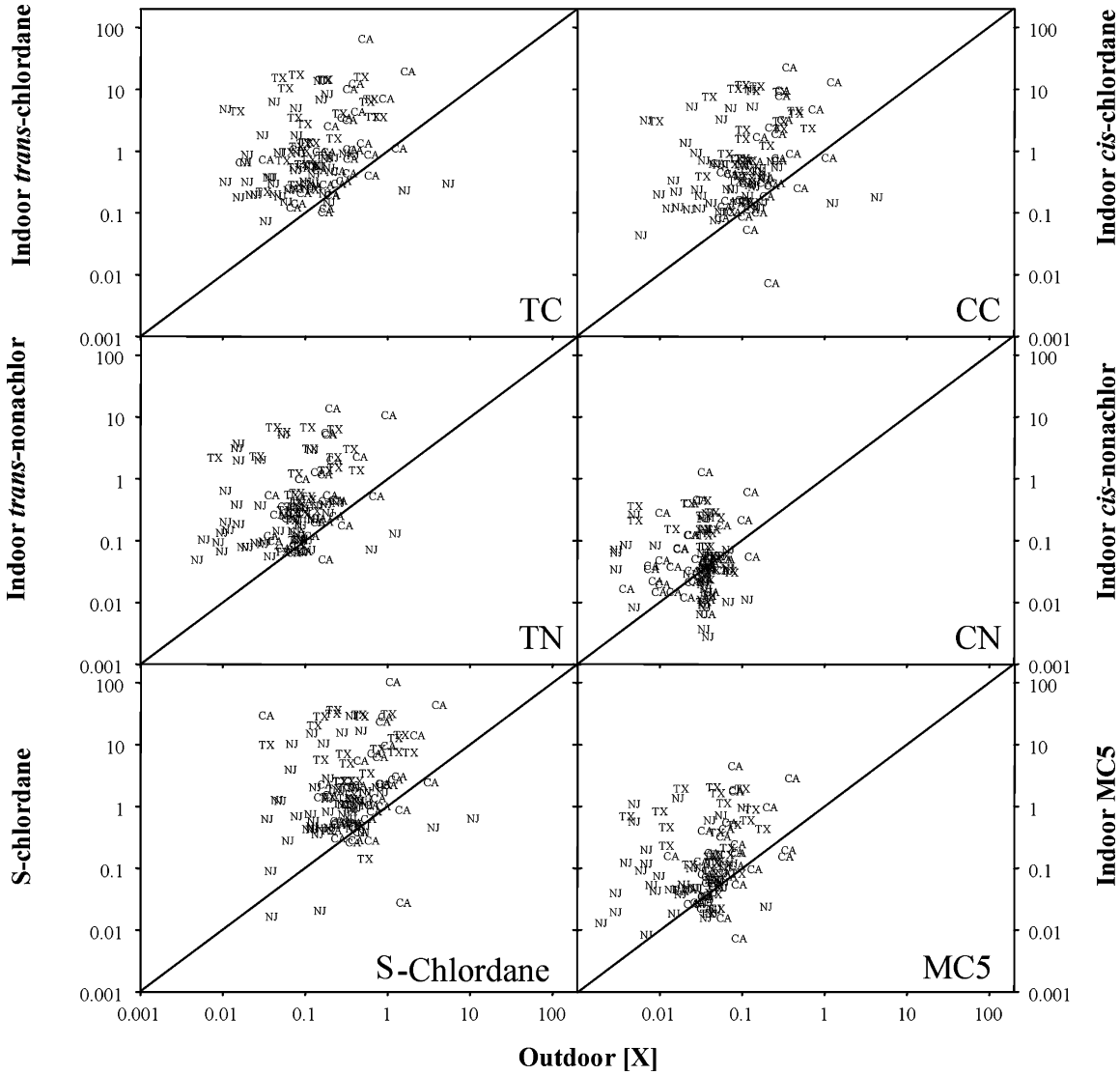

FIGURE 3. Plot of indoor vs outdoor concentrations (ng m^{-3}) of individual total (gas + particle) chlordane species (*trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, *cis*-nonachlor, MC5) and Σ-chlordane in paired samples from Elizabeth, NJ, Los Angeles County, CA, and Houston, TX. Note log−log scaling of axes.

where α is the air exchange rate (h^{-1}), P is the penetration factor (ranging from 0 to 1), k is the loss rate constant (h^{-1}), C_{in} and C_{out} are total (gas + particle phase) concentrations measured indoors and outdoors (ng/m^3), S is the indoor source strength (ng h^{-1}), and V = total home air volume (m^3).

At steady state ($dC_{in}/dt = 0$), eq 1 can be expressed as

$$C_{in} = [P\alpha/(\alpha + k)]C_{out} + S/V(\alpha + k) \qquad (2)$$

where the first term on the right-hand side of the equation is the outdoor contribution to the indoor concentrations and the second is the corresponding contribution of indoor sources expressed in ng m^{-3}. Lower bound indoor emission rates were estimated for each home using measured indoor and outdoor concentrations, air exchange rates, and house volumes. For those samples in which one phase was below the limit of detection, the MDL was used in these calculations. Air exchange rates were measured for each home and are described in greater detail by Meng et al. (*63*). As a limiting case, the penetration factor was assigned a value of unity, and the reaction rate constant was set to zero. Ambient outdoor half-lives for chlordane species are on the order of ~7 to ~10 days, which translate into reaction rate constants of approximately 0.003 h^{-1} (*71*, *72*). Also, since ~90% of airborne chlordanes are typically found in the gas phase, the following typical assumptions for nonpolar gases are likely a good first approximation for calculating a lower bound limit for the indoor source strength in each home for which paired indoor and outdoor (gas + particle) samples are available.

When $P = 1$ and $k = 0$, eq 2 becomes

$$C_{in} = C_{out} + S/V\alpha \qquad (3)$$

where C_{out} also represents the outdoor contribution to the observed indoor concentrations in ng m^{-3}.

In this way, a limiting lower boundary for indoor source strength can be calculated for each home for which paired indoor and outdoor (gas + particle) concentrations, home volumes, and air exchange rates are available (n = 112). Lower-bound indoor source strengths (S) average 1520 ± 3290 ng h^{-1} (geometric mean: 326 ng h^{-1}; maximum: 17 500 ng h^{-1}; minimum: −965 ng h^{-1}). Large, positive source strengths indicate an indoor source (or sources) of chlordanes. Calculated indoor source strengths were positive for 100 of the 112 homes (geomean: 326 ng h^{-1}; geometric mean +1 SD: 2750 ng h^{-1}; geometric mean −1 SD: 38.7 ng h^{-1}),

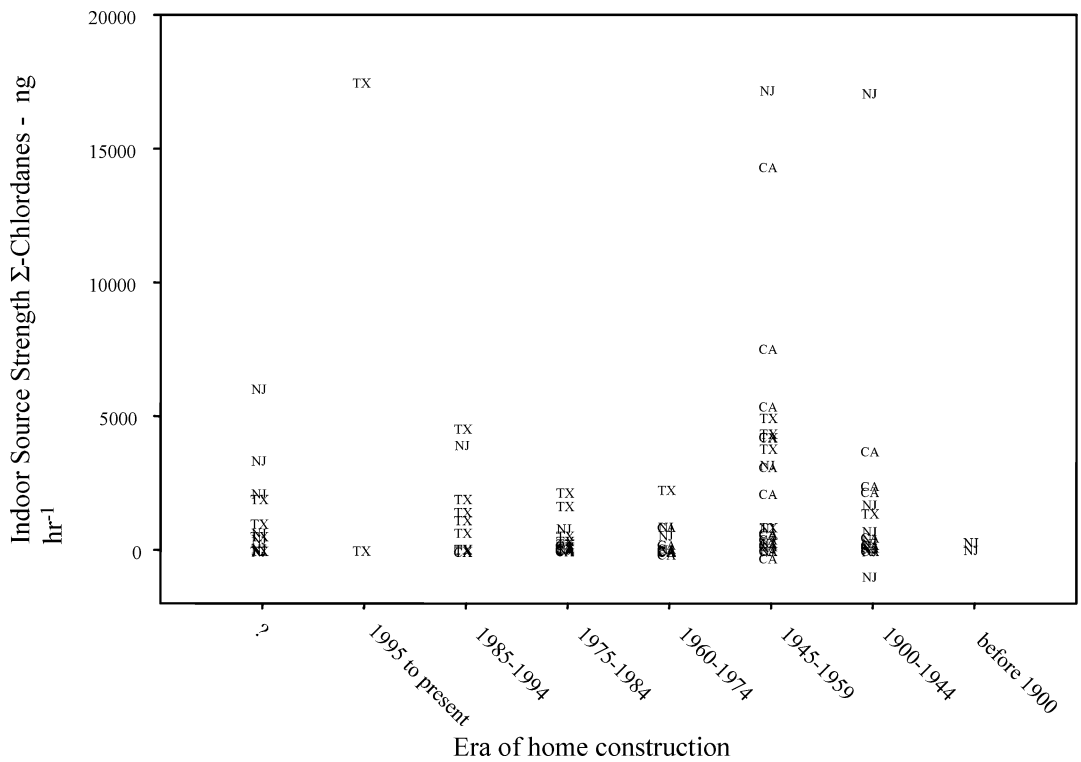

FIGURE 4. Calculated source strengths of total (gas + particle) Σ-chlordane (TC + CC + TN + CN) in Elizabeth, NJ, Los Angeles County, CA, and Houston, TX.

and 24 of these homes have calculated Σ-chlordanes source strengths greater than 2000 ng h^{-1}. Indoor sources of chlordanes likely include volatilization from residues of indoor application of chlordanes and infiltration from subsurface and foundation application of chlordane-containing termiticides during home construction. The lower bound geometric mean calculated indoor emission rate of 326 ng h^{-1} translates into a total mass flux of 0.113 g over a period of 40 years, assuming constant emission. This calculated emission flux is several orders of magnitude lower than the typical application of several kilograms and indicates that strong indoor emissions may continue for many years.

The date of home construction appears to play a significant role in indoor concentrations of chlordanes in all three U.S. cities, while there is no apparent dependence of calculated source strengths on temperature or air exchange rate. Five of the eight highest emission rates of total Σ-chlordanes (all >5000 ng h^{-1}) occur in homes that were constructed during the period from 1945 to 1959 (Figure 4). All eight of these homes are detached (i.e. unattached to another structure) single family homes. With the exception of one Texas home-built during the period from 1985 to 1994 and one New Jersey home with an unknown construction date, all 24 homes that have apparent emission rates greater than 2000 ng h^{-1} are detached single family dwellings. The two exceptions are single family homes that are attached to another structure on a single side.

Outdoor Contribution to Indoor Concentrations. Following the above assumptions of perfect penetration ($P = 1$), of negligible loss rate ($k = 0$) and of only two possible source zones (indoors or outdoors), the upper bound relative outdoor contribution (ROC) to the observed indoor concentration, expressed as a percent, can be calculated as follows

$$\mathrm{ROC}_i = C_{\mathrm{out},i}/C_{\mathrm{in},i} \times 100 \qquad (4)$$

where C_{in} and C_{out} are the indoor and outdoor concentrations of chlordane species i, respectively. Thus, the lower-bound relative indoor contribution (RIC) equals 100-ROC (i.e. RIC + ROC = 100). For simplicity, only those homes with a RIC greater than 0.1% are included in the following analysis. Thus, homes for which the lower bound value of RIC was negative, or not largely different from zero, have been removed from the following analysis. For example, the RIC for OXY is greater than 0 in 39 of 84 homes (or 46.4% of the 84 homes), which means that 45 homes (53.6% of the 84) had an indoor source contribution not greater than 0%, using the assumptions of $P = 1$ and $k = 0$.

TABLE 4. Upper Limit Relative Outdoor Contribution (ROC) to Indoor Concentration (C_{in}) by Compound Where ROC = 100 − RIC, Where RIC = $\{C_{\mathrm{in}} - C_{\mathrm{out}}/C_{\mathrm{in}}\} \times 100$, Using the Assumptions of $P = 1$ and $k = 0$ in Eq 2 in the Text

	OXY	TC	CC	MC5	TN	CN	Σ-chlor
N	39	103	100	93	102	67	112
mean	76.0%	20.0%	24.2%	35.2%	30.1%	39.6%	25.8%
SD	22.3%	19.7%	22.7%	29.6%	27.4%	31.9%	23.3%
geometric mean	86.0%	23.5%	30.3%	47.9%	40.1%	54.5%	32.7%
median	83.8%	12.9%	15.4%	28.6%	21.5%	26.7%	20.3%
min	5.1%	0.2%	0.2%	0.4%	0.3%	1.3%	0.1%
max	98.7%	81.4%	98.6%	97.7%	97.2%	97.4%	99.2%
%w/RIC > 0.1%	46.4%	92.0%	89.3%	83.0%	91.1%	62.0%	89.6%
no. of homes	84	112	112	112	112	108	125

A summary of the resulting relative contributions of outdoor chlordane concentrations to the observed indoor chlordane concentrations is given in Table 4. The Σ-chlordane concentration exhibits a geometric mean upper bound ROC of 32.7%, which indicates that for 112 of 125 homes for which this calculation gave a result for RIC greater than zero, approximately one-third of the observed concentration of Σ-chlordanes at indoor locations was from outdoor penetration into the indoor environment. This means that, for the "average home" in this study, at least two-thirds of the observed concentrations comes from indoor sources.

On an individual compound basis, upper bound geometric mean ROCs range from 54.5% for *cis*-nonachlor to 23.5% for

trans-chlordane with the exception of oxychlordane (83.8%). Those compounds that are most prominent in technical chlordane, as it was applied (TC, CC, TN), are those that have the lowest relative outdoor contributions, while OXY appears to behave differently from the others, showing the greatest relative outdoor importance to observed indoor concentrations. This indicates that indoor concentrations of OXY are not dominated by formation indoors, although the calculations described above indicate that formation of OXY at indoor locations may occur in approximately half of the homes (39 of 84 homes) for which paired indoor and outdoor concentrations of OXY were measured.

Importance of Indoor Sources to U.S. Ambient Atmospheric Burden. Assuming the geometric mean indoor source strength of Σ-chlordane (326 ng h^{-1}), as calculated above, is representative of homes across the entire country, and that there are ~100 million homes across the United States, then the national annual flux from residential, indoor sources would be ~300 kg yr^{-1} or ~0.8 kg day^{-1}. This calculation assumes that all of the chlordane emitted by indoor sources, as calculated above, escapes from the homes. Thus, this simplified calculation represents an estimate of the importance of indoor emissions to the outdoor atmosphere.

The annually averaged, total atmospheric burden in the surface-mixed layer ($h = 1000$ m) over the 48 contiguous states of the United States (*73*) is ~800 kg on an "average day" (assuming a concentration of 0.1 ng m^{-3}: thus 0.1 ng m^{-3} * 1×10^{-12} kg ng^{-1} * 1×10^{9} m^3 km^{-3} * 7 841 730 km^2 * 1000 m * 1 km/1000 m = 800 kg). Therefore, the daily loading from indoor residential sources represents ~0.1% of the surface mixed layer burden. This suggests that release from homes does not play a major role as a source of chlordanes input into the ambient atmosphere in the United States. Furthermore, this suggests that emission from residences in the continental Unites States likely does not constitute an appreciable source of these compounds to either the Arctic or Antarctic burdens. Eitzer et al. (*74*) have suggested that residential (nonagricultural) sources may play an important role in observed ambient air concentrations in Connecticut. The data presented here do not provide strong evidence that indoor sources greatly enrich ambient atmospheric concentrations. It is important to note that this does not exclude the possibility that volatilization from areas of former outdoor residential use may indeed provide a substantial source of these compounds to ambient air.

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Supporting Information Available

Information about analysis, summary descriptive statistics of measured chlordane concentrations, and the measured concentrations of 6 chlordane compounds (gas and particle bound) in the indoor and outdoor air in Los Angeles County, CA, Houston, TX, and Elizabeth, NJ homes. This material is available free of charge via the Internet at http://pubs.acs.org.

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