

ORIGINAL RESEARCH

Aggregate exposures of nine preschool children to persistent organic pollutants at day care and at home

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In the summer of 1997, we measured the aggregate exposures of nine preschool children, aged 2–5 years, to a suite of organic pesticides and other persistent organic pollutants that are commonly found in the home and school environment. The children attended either of two child day care centers in the Raleigh–Durham–Chapel Hill area of North Carolina and were in day care at least 25 h/week. Over a 48-h period, we sampled indoor and outdoor air, play area soil and floor dust, as well as duplicate diets, hand surface wipes, and urine for each child at day care and at home. Our target analytes were several polycyclic aromatic hydrocarbons (PAH), organochlorine pesticides, and polychlorinated biphenyls (PCB); two organophosphate pesticides (chlorpyrifos and diazinon), the lawn herbicide 2,4-dichlorophenoxyacetic acid (2,4-D), three phenols (pentachlorophenol (PCP), nonyl phenols, and bisphenol-A), 3,5,6-trichloro-2-pyridinol (TCP), and two phthalate esters (benzylbutyl and dibutyl phthalate). In urine, our target analytes were hydroxy-PAH, TCP, 2,4-D, and PCP. To allow estimation of each child's aggregate exposures over the 48-h sampling period, we also used time–activity diaries, which were filled out by each child's teacher at day care and the parent or other primary caregiver at home. In addition, we collected detailed household information that related to potential sources of exposure, such as pesticide use or smoking habits, through questionnaires and field observation. We found that the indoor exposures were greater than those outdoors, that exposures at day care and at home were of similar magnitudes, and that diet contributed greatly to the exposures. The children's potential aggregate doses, calculated from our data, were generally well below established reference doses (RfDs) for those compounds for which RfDs are available.

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Introduction

In previous work in the early spring of 1997, we measured the concentrations of several persistent organic pollutants (POP) in 10 child day-care centers in the Raleigh–Durham–Chapel Hill area of North Carolina (Wilson et al., 1999, 2001). From these centers, we chose the two at which we had found the overall highest or the overall lowest concentrations of the targeted compounds. Then in the summer of the same year, we measured the aggregate exposures of nine preschool children, ages 2–5 years, from the chosen two centers, to the same suite of organic pesticides and other POP that are commonly found in the home and school environment. The targeted pollutants were chosen because they often persist indoors and outdoors; they are distributed widely (Wania and

MacKay, 1996); they have been used in schools and homes; and they can result in acute or chronic toxicity at high levels of exposure. These pollutants included polycyclic aromatic hydrocarbons (PAH); benzylbutyl and dibutyl phthalate; the organophosphate pesticides diazinon and chlorpyrifos (CPF); organochlorine pesticides such as DDT, chlordane, and heptachlor; polychlorinated biphenyls (PCBs); pentachlorophenol (PCP), nonyl phenols, and bisphenol-A; and the lawn herbicide 2,4-dichlorophenoxyacetic acid (2,4-D).

Young children are considered to be especially susceptible to the harmful effects of pollutants both because their bodies are immature and developing rapidly, and because their behavior, such as testing new objects with their mouths, may increase their exposures (Zahm and Devesa, 1995; Heil et al., 1996; Gurunathan et al., 1998; Mukerjee, 1998; Landrigan et al., 1999; Cohen Hubal et al., 2000a, b; Freeman et al., 2001a, b; Tulve et al., 2002). Children's exposures can come from several media, including air, dust, soil, and food. The routes of the exposures may be inhalation (Ny et al., 1993; EFH, 1995), dietary and nondietary ingestion (Waldman et al., 1991; Menzie et al., 1992; Stanek and Calabrese,

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1995a, b; Lewis et al., 1999; Curl et al., 2002; Fenske et al., 2002a, b), and dermal absorption (Mukerjee, 1998; Griffin et al., 1999; Lu and Fenske, 1999).

In this study, we measured the targeted compounds in several environmental media: indoor and outdoor air, house and classroom floor dust, and outdoor play area soil. We also measured the targeted compounds in several personal media: duplicate diet solid and liquid food, hand wipes, and urine. We obtained time-activity diaries and household observational data for all the children. Our objectives were to evaluate and refine methods for the study of preschool children's exposures, to measure the concentrations of the targeted POP in multiple media at children's homes and day-care centers, to estimate the range of concentrations and the children's aggregate exposures to and potential doses of the targeted pollutants, to examine possible differences between exposures at home and at day care, and to acquire information to aid us in the design of a larger study of preschool children's exposures, (Wilson et al., 2000, 2001, 2002; ORD, 2000).

Based on our findings in our preceding study of multi-media concentrations of these pollutants at 10 child day-care centers in the same geographic area of North Carolina (Wilson et al., 2001), we formulated three hypotheses to evaluate in this study and for future testing: (1) Young children derive a large part of their exposures to, and potential doses of, POP through the ingestion pathway. (2) The children's exposures at home and at day care are similar. (3) The exposures of children in low-income families and middle-income families are similar.

The participants in this study were nine children who attend child day-care centers at least 25 h/week. The children ranged in age from 2 to 5 years; their average age was 4. Five of the children were from a center that serves primarily middle-income families, and four were from a center that serves primarily low-income families. One child was Hispanic, three were African American, and five were Caucasian. During the sampling period, they spent about 40% of their time at day care and most of the remaining time at home.

Methods and materials

Selection of Participants

From 10 child day-care centers that participated in our previous study (Wilson et al., 2001), the two centers were chosen that exhibited the lowest and the highest overall levels of our target pollutants. One center is located within the city limits of Durham, North Carolina; the other is located in a small, primarily rural community outside Raleigh, North Carolina. With the permission of the center directors, introductory letters were sent to all the parents of preschool children ages 2-5 years at each center. These letters included

a participation summary sheet and a brief home survey, asking the parents to contact us for further information if they were interested in participating. Since only two parents were interested initially, new simplified flyers were distributed, with the permission of the center directors, to parents of children in the appropriate classrooms. Information tables were set up outside the centers, where parents could talk with us and ask questions about the study. There were 66 eligible children, ages 2-5 years, who were toilet-trained, and who attended either of the two centers. In all, 11 families were recruited successfully, of whom nine families actually participated in the study.

Sampling

All the sampling was conducted over a 2-week period. In the first week, sampling took place at one day-care center and at the homes of four children who attended that center. Two of the children, who were in the same classroom, participated in the first 48-h period, and the other two children, who were in a second classroom, participated in the second 48-h period that week. Similarly, in the second week, sampling took place at the second center and at the homes of five children who attended that center. Three children at the second center were in one classroom, and two were in another.

Sampling over the 48-h period was conducted simultaneously at each child's day-care center and at the child's home. Indoor and outdoor air, floor dust, play area soil, and duplicate diet samples were collected similarly to our earlier study of ten day-care centers (Wilson et al., 2001). In addition, hand wipes and urine samples were collected from each child. The children's parents and teachers were trained by our staff to collect the diet, hand-wipe, and urine samples. Additionally, these caregivers filled out time-activity diaries for each child. The diaries included time, place, surface, activity, and duration. Household questionnaires, covering family characteristics, occupation, income, education, use of pesticides, typical eating habits, type of cooking, etc., were administered by our field staff, who also gathered observational data on house and child day-care center characteristics, such as traffic density, location near possible pollutant sources, industries, etc.

Indoor and outdoor air at day care and at home were sampled for 48 h at 4 l/min with a URG sampler (University Research Glassware Corp., Chapel Hill, NC, USA). The 10 μ m impactor-equipped inlet was followed by a glass cartridge containing a quartz fiber filter followed by XAD-2 resin for the targeted compounds except 2,4-D, or containing a Teflon-coated glass fiber filter followed by polyurethane foam (PUF) for 2,4-D, to collect both particulate- and vapor-phase materials (Chuang et al., 1987, 1991, 1999b, c; Wilson et al., 1991, 1996). The indoor sampler was placed in a Styrofoam box equipped with a cooling fan, and then in a playpen with a net over the top to protect the equipment

from curious children. The outdoor sampling pump and controls were sheltered in a dog house, which we provided, located centrally in the children's outdoor play areas. The air sampler inlets were placed at the approximate breathing height of the children, about 75 cm from the floor or ground.

Classroom and house floor dust were collected with a High Volume Small Surface Sampler (HVS3; Cascade Stack Sampling Systems Inc., Bend, OR, USA) using a standard ASTM method (Chuang et al., 1994; Lewis et al., 1994; ASTM, 1997) in the areas indicated by the teacher or parent as being where the children played most often. Play area soil was scraped from the top 0.5 cm of exposed soil over a 0.1 m² (1 ft²) area in the location identified by the teacher or the parent as being most used by the children (Chuang et al., 1995). The selected outdoor play areas generally had little or no grass.

Duplicate plates (Acheson et al., 1980; Block, 1982; Fennema and Anderson, 1991) of the daily meals served to the children in the participating classrooms and at home were collected on each of the two sampling days in the 48-h period. The same menus were served to all the children in a given classroom. Menus at home varied considerably, as expected. If a child brought his or her food from home, rather than eating food served at the day care, a duplicate of this food from home was collected. The foods were collected each day as composites of food eaten at home, and composites of food eaten at day care. Liquid and solid foods were collected separately in glass containers (Chuang et al., 1998; Wilson et al., 2001) and refrigerated until they were shipped on dry ice to the laboratory for analysis.

Each child's hand-wipe samples were collected by the child's caregiver. The wipes were of cotton gauze (Johnson and Johnson Sof-Wick). They were precleaned with dichloromethane (DCM), dried, then wetted before use with 2 ml of a 50% isopropanol/water solution. Two wipes for each child were collected at the day-care center, one just before lunch and before washing the child's hands, on each of the two sampling days. Two additional wipes were collected at home, just before dinner and before washing the child's hands, on each of the two sampling days.

Each child's urine samples were collected using a plastic bonnet placed under the toilet seat, then transferred to a plastic bottle by the child's caregiver. Four urine samples were collected at home: one in the morning (the first morning void) and one in the evening before bedtime on each sampling day. Two urine samples were collected at day care, after lunch on each sampling day.

The wipe and urine samples were stored in chilled coolers at the centers or at home until collected by our staff. They were then stored in freezers until they were shipped on dry ice to the laboratory, then stored in freezers (-20°C) until analyzed.

At least one field blank sample was collected in each sampled medium at each location. Additional details of the

sampling and analytical methods are published elsewhere (Chuang et al., 1998).

Analysis

The target compounds for analysis included 20 PAH, seven of which are classified as probable human carcinogens, B2 PAH; (IRIS, 1999; IARC), two phthalate esters (PE), two organophosphate pesticides (OP), nine organochlorine pesticides (OC), 18 PCBs, three phenols (Ph), the lawn herbicide 2,4-D, and the CPF metabolite 3,5,6-trichloro-2-pyridinol (TCP).

The filter/XAD-2 air samples and field blanks were spiked with surrogate recovery standards (SRS): pyrene-d₁₀, chrysene-d₁₂, DDE-¹³C, DDT-¹³C, 2,2',4,5,5'-pentachlorobiphenyl, and fenchlorfos, and Soxhlet-extracted with DCM. The DCM extract was concentrated by Kuderna-Danish evaporation, and divided into two portions. The first portion was analyzed by gas chromatography/mass spectrometry (GC/MS) operated in the electron impact (EI), selected ion monitoring (SIM) mode, for PAH, PCB, PE, Ph, and OC and OP pesticides. Three GC/MS analyses were performed for each extract: one for PAH, Ph and PE, one for PCB, and one for OC and OP.

Only the filter of the filter/PUF air samples was analyzed for 2,4-D, since our method evaluation tests (Chuang et al., 1998) showed that the 2,4-D is retained in the filter. After spiking with the SRS 3,4-dichlorophenoxyacetic acid (3,4-D), the filter was extracted sonically with a phosphate buffer solution, and the extract was processed through a C18 solid-phase extraction (SPE) column. The target fraction was concentrated, methylated, and analyzed by GC with electron capture detection (ECD) for 2,4-D.

Dust samples were separated into coarse and fine (<150 μm) fractions. Literature reports indicate that only the fine fraction of house dust contains significant quantities of the targeted organic pollutants (Lewis et al., 1999). Additionally, the fine fraction is most likely to adhere to and remain on human skin (Duggan et al., 1985; Que Hee et al., 1985; Driver et al., 1989). Therefore, only the fine dust fraction was analyzed. Soil samples were not separated into fractions (Lewis et al., 1999). Both dust and soil were spiked with SRS and extracted twice each sonically with 10 ml of 10% diethyl ether (EE) in hexane for 20 min. The extracts were concentrated to 1 ml and applied to a Florisil SPE column, preconditioned with 50% EE in hexane followed by 100% hexane. The fraction eluted with 12 ml 15% EE in hexane was concentrated and analyzed by GC/MS for PAH, PCB, PE, OC, and OP.

A second aliquot of each dust and soil sample was used for analysis of Ph. The samples were extracted sequentially with 5% acetic acid in methanol, DCM, and 5% acetic acid in distilled water. The mixture was washed with distilled water, and the DCM extract was concentrated. One portion of the extract was methylated and analyzed for PCP and the other

portion was not methylated and was analyzed for the other Ph.

A third dust/soil aliquot was extracted with phosphate buffer, cleaned up with C18 SPE, and analyzed for 2,4-D by GC/ECD (soil) or GC/MS (dust).

A fourth aliquot of dust/soil was extracted with acetonitrile (ACN) at 120°C under 2000 psi using accelerated solvent extraction (ASE). The ACN extract was solvent-exchanged into toluene; derivatized with *N*-methyl-*N*-(*t*-butyldimethylsilyl)-trifluoroacetamide (MTBSTFA); and analyzed by GC/MS for TCP.

The solid and liquid food samples were extracted separately (to allow comparison of these media as sources of exposure) and analyzed for the target compounds. Aliquots of the homogenized food samples were spiked with SRS, extracted with DCM, fractionated by gel permeation chromatography (GPC), and split into two portions. One portion was methylated and cleaned up with Florisil SPE for the determination of 2,4-D and PCP. The second portion was not methylated and was cleaned up with Florisil SPE for determination of the other target compounds. All the fractions were concentrated and analyzed by GC/MS.

A second aliquot of solid and liquid food samples was extracted with ACN at 100°C under 2000 psi using ASE. The resulting ACN extract was concentrated to 1 ml and diluted with 25 ml of acetone. The acetone extract was processed for acid–base partitioning and derivatization with MTBSTFA prior to GC/MS analysis for TCP.

The hand-wipe samples collected at home for each child were extracted together, as were the hand-wipe samples collected at day care. After addition of SRS, each combined sample was Soxhlet-extracted with 10% EE in hexane. The extract was dried with anhydrous sodium sulfate, concentrated to 1 ml, applied to a preconditioned Florisil SPE column, and eluted with 24 ml 15% EE in hexane. After concentration, this fraction was analyzed by GC/MS. The hand-wipe samples were not analyzed for 2,4-D, which would have required additional samples and a different analytical method.

For each subject, the urine samples collected at home were combined, as were the urine samples collected at day care. One 30-ml aliquot of each composite sample was used for determining hydroxy-PAH, PCP, and 2,4-D. A 1-ml aliquot was used for determining TCP. A 5-ml aliquot was sent for creatinine analysis (Smith Kline Beecham Laboratories, St Louis, MO, USA).

Urine samples were analyzed for hydroxy-PAH, PCP, and 2,4-D by hydrolysis with 6 N HCl, methylation with diazomethane, fractionation with a Florisil SPE column, and GC/MS. The samples were analyzed for TCP by hydrolysis with 6 N HCl, extraction with DCM, solvent exchange from DCM to toluene, derivatization with MTBSTFA, and GC/MS.

All GC/MS analyses of sample extracts and standard solutions were done on a Hewlett-Packard GC/MS, operated in SIM mode (70 eV EI), with a ChemStation data system. The GC column was a DB-5 fused silica capillary column (60 m × 0.32 mm; 0.25 μm film thickness), used with helium carrier gas. Following injection, the GC column was held at 70°C for 2 min, then programmed to 150°C at 15°C/min, then to 290°C at 6°C/min. Molecular ions and their associated characteristic fragment peaks were monitored. Identifications were based on the GC retention times relative to those of corresponding internal standards and the relative abundances of the monitored ions. Quantifications were based on comparisons of the integrated ion current responses of the target ions to those of the corresponding internal standards using average response factors of the target analytes generated from standard calibrations.

Overall, 55 different chemical compounds were measured in eight media. The limits of detection (LODs) were estimated from the analytes' signal-to-noise ratios at the lowest levels of standard calibrations. These LODs for the various compound classes in the sampled media are given in Table 1. Additional details of the methods and method validations for this study are available elsewhere (Chuang et al., 1998).

Statistical Assessments

Statistical assessments were conducted on the following sample types: 48-h integrated indoor and outdoor air, HVS3 house and classroom dust, play area soil, 48-h duplicate diet, composite hand-wipe, and composite urine samples. Summary statistics (sample size, mean, median, standard deviation, minimum, and maximum) were determined for each of the preceding sample types. Four additional types of statistical analysis were performed: Spearman correlation analyses, Pearson correlation analyses, analysis of variance (ANOVA), and regression models. Summary statistics and Spearman analyses were done on the raw data, while the remaining analyses were done on natural log-transformed data.

Summary statistics were generated for the measured data by compound, sample medium, and family income. The low-income households were categorized using the federal guidelines for assistance under the Women, Infants, and Children (WIC, 1997), that is, household income less than 185% of the federal poverty guidelines. Summary statistics were also generated on the calculated estimated daily doses of the POP by compound, exposure pathway, and family income.

Spearman and Pearson correlation coefficients were calculated to examine the relations between POP concentrations in different sample media, and to examine the relations of the POP concentrations in each compound class within the sample medium.

An ANOVA model was used to examine the effects of sampling location (home vs. day-care center) and family

Table 1. Estimated limits of detection for the target analytes in multimedia samples.

Compound class	Air (ng/m ³)	Dust/soil ppm (μg/g)	Liquid food ppb (ng/g)	Solid food ppb (ng/g)	Hand wipes (ng/wipe)	Urine (ng/ml)	Urine (μmol/mol)
Total PAH	0.01–0.04	0.001	0.04	0.04	0.5	NA ^a	NA
B2 PAH	0.01–0.04	0.001	0.04	0.04	0.5	NA	NA
Phthalates	0.04	0.001	0.04	0.04	0.5	NA	NA
OP pesticides	0.1	0.001	0.04	0.04	0.5	NA	NA
OC pesticides	0.1	0.001	0.04	0.04	0.5	NA	NA
PCB	0.04	0.001	0.04	0.04	0.5	NA	NA
Phenols	0.1	0.001	0.1	0.04	0.5	NAP ^b	NAP ^b
2,4-D	0.1	0.001	0.04	0.5	0.5	0.03	0.64–2.77 ^c
Hydroxy-PAH	NA	NA	NA	NA	NA	0.017	0.005–0.073 ^c
TCP	NA	0.001	0.05	0.5	NA	1.0	0.64–2.79 ^c
PCP	0.1	0.001	0.1	0.04	0.5	0.1	0.48–2.11 ^c

^aNA indicates that the samples were not analyzed for this analyte.

^bNAP indicates that these samples were analyzed only for pentachlorophenol (PCP).

^cAdjusted to micromoles per mole creatinine. The detection limit in μmol/mol varied from sample to sample; the ranges are reported.

income (low- vs. middle/upper-income) on POP concentrations for each sample medium. The natural log-transformed data in multimedia samples (air, dust, soil, food, wipe, and urine) were used for ANOVA. Urine data, calculated in two units — the weight per unit volume, ng/ml (not creatinine-adjusted) and μmol/mol (creatinine-adjusted) — were fitted separately to the ANOVA models.

Regression models were used to examine the relations between urinary metabolite concentrations and POP concentrations in the sampled media, as well as between urinary metabolites and estimated total daily POP dose. Two different types of regression models were fitted to the urinary metabolite data. The first type of model included factors for sampling location (day care vs. home) and POP concentrations. The POP data in play area soil were not included in the model, because these concentrations in soil were low, and initial analyses as well as literature reports (Fenske et al., 2000) showed that soil was not a significant factor for the urinary metabolite concentrations. The second type of model included factors for sampling location and estimated aggregate daily POP doses. The estimated aggregate daily POP doses were computed as the sums of the estimated daily doses from inhalation, nondietary ingestion, and dietary ingestion that resulted from the exposures at homes and at day-care centers. Separate models were fitted to selected hydroxy-PAH, PCP, 2,4-D, and TCP data in subjects' urine samples. All regression models were fitted to the ln-transformed data.

Estimating Exposures and Potential Dose

The potential daily dose for a given child is calculated from the aggregate daily exposure of the child, where the aggregate exposure is defined as the total weight of a pollutant to which the child is exposed daily, in ng/day, through contact with all media. The potential dose is then the total exposure, through

all routes and media, per kg body weight of the child per day. Assuming 100% absorption, the potential dose gives upper limits on the amount of the compound available for delivery to target organs. Since the concentrations of the targeted compounds found in this study were generally low, and the targeted compounds were not associated with fresh applications, the subject children's dermal exposures are expected to be low. Literature reports support this expectation. For example, in one study, less than 1% of CPF from a formulation applied to environmental surfaces 3.5 h earlier was transferred to skin surfaces (Lu and Fenske, 1999). In another study, in which whole-body dosimeters were used, approximately 1.3 mg/kg per person transferred from CPF-treated turf, but only 1.3 μg/kg per person was absorbed, based on urinary TCP values (Bernard et al., 2001). Of a relatively large amount (28.59 mg) of CPF applied to the skin of five adult human volunteers and left there for 8 h, only 1% of the applied dose was recovered in urine, with an implied half-life of 30 h (Griffin et al., 1999). Therefore, the contribution of dermal absorption to exposure is expected to be negligible compared to the contributions from other media in this study and is not included in our calculations.

The potential daily dose of each POP in μg/kg per day was calculated using the following three equations. The first equation expresses the maximum daily dose through inhalation as

$$D_{inh} = (C_i t_i + C_o t_o) VF / 1000 W (t_i + t_o)$$

where D_{inh} is the estimated dose through inhalation at home or at day care in μg/kg/day, C_i is the indoor air POP concentration at home or at day care in ng/m³, C_o is the outdoor air POP concentration at home or at day care in ng/m³, t_i is the time the child spends indoors at home or at day care in min, t_o is the time the child spends outdoors at home

or at day care in min, V is the child's estimated ventilation rate (the total volume of air inhaled per day), which is assumed to be $8.3\text{ m}^3/\text{day}$ (EFH, 1995), W is the measured body weight of the child in kg, and F is the fraction of the 24-h day that the child spent at home or at day care.

The maximum daily dose through nondietary ingestion is based on the dust and soil concentrations, the amounts of time indoors and outdoors, and the time that the child spends at home and at day care, which is expressed as

$$D_n = (D_i t_i + P_o t_o) M F / W (t_i + t_o)$$

where D_n is the estimated dose through nondietary exposure at home or at day care in $\mu\text{g}/\text{kg}\text{ day}$, D_i is the POP concentration in floor dust at home or at day care in $\mu\text{g}/\text{g}$, P_o is the POP concentration in the pathway soil at home or at day care in $\mu\text{g}/\text{g}$, and M is the weight of dust/soil ingested by the subject daily, which is assumed to be $0.1\text{ g}/\text{day}$ (Stanek and Calabrese, 1995a, b).

The maximum daily dose through dietary ingestion is based on the POP concentrations in the liquid and solid food composite samples and the mass of the food samples, which is expressed as

$$D_d = (S_i M_s + L_i M_l) / 2W$$

where D_d is the estimated dose through dietary exposure at home or at day care in $\mu\text{g}/\text{kg}\text{ day}$, S_i is the POP concentration in the solid food samples at home or at day care in $\mu\text{g}/\text{kg}$, M_s is the total weight in kg of solid food intake by the subject in the 48-h period, L_i is the POP concentration in the liquid food samples in $\mu\text{g}/\text{kg}$, and M_l is the total weight of liquid food intake by the subject in the 48-h period.

Results and discussion

Concentrations

The concentrations of the individual target compounds at home and at day care are given in Tables 2–5. Tables 2 and 3 show the mean concentrations and ranges of concentrations of the compounds in air, floor dust, and soil at the two child day-care centers and at the children's homes. The numbers of samples below the LOD for each compound are given in parentheses after the range of concentrations. In calculation of the mean concentrations, one-half the LOD was used for those samples below the LOD, which may cause the calculated means to be slightly higher than they might have been if the targeted pollutants were simply not present instead of being at levels too low to detect. The herbicide 2,4-D was seldom detected in indoor or outdoor air at homes and at centers. All the other target compound classes were detected in most of the analyzed air samples. The mean indoor air concentrations of all the target compounds were higher than those in outdoor air. Many of the target compounds were not detected at all in the soil samples, and

most of the concentrations in soil were lower than those in floor dust.

Tables 4 and 5 show the mean concentrations and ranges of concentrations of the compounds in the children's food and hand wipes, at home and at day care. In liquid food, concentrations of most of the target compounds, including most of the PAH, the OP and OC pesticides, the PCBs, and the Ph, were near or below the LOD. Benzylbutylphthalate, 2,4-D, and TCP were found in all of the liquid food samples. In solid food, concentrations of the larger B2 PAH, the OC pesticides, the PCBs, PCP, and 2,4-D, were also near or below the LOD. The two target phthalates; the PAH naphthalene, fluorene, fluoranthene, and pyrene; CPF; nonyl phenols; bisphenol-A; and TCP were found in all solid food samples.

The target compounds were seldom found in the composite hand-wipe samples, either at day care or at home. However, the range of concentrations in the hand wipes was large, except for the PCBs, which ranged only from less than to about three times the LOD.

Table 6 shows the concentrations of the target compounds in the children's urine samples, composited over the entire 48-h sampling period for each child at home and day care. The actual concentrations are given in ng/ml , as well as the concentrations that have been normalized to $\mu\text{moles per mole}$ of creatinine. Although creatinine normalization is used frequently, to account for variability in dilution of the urine, in this study the creatinine-normalized values showed poorer correspondence with the measured exposures than did the non-normalized values. This supports the reported caution against using creatinine normalization in evaluating doses for children (O'Rourke et al., 2000).

The benzo[a]pyrene metabolites, 1- and 3-hydroxybenzo[a]pyrene, were detected in only two of the nine composite urine samples. The PAH metabolites 1- and 2-naphthol, 3-hydroxyfluoranthene, 1-hydroxypyrene, and 6-hydroxy-chrysene, as well as PCP, 2,4-D, and TCP, were found in all urine samples.

Statistical Assessments

Sampling location, day care vs. home, exhibited statistically significant differences. Home levels of the targeted compound classes were higher than those at the centers for OP pesticides in indoor air ($P < 0.05$). They were lower than those at the centers for total PAH in indoor air, PCB in indoor air ($P < 0.05$) and outdoor air ($P < 0.01$), and Ph in liquid food and floor dust ($P < 0.05$).

The OP pesticide concentrations in liquid food were higher for the children in low-income families, compared to those in middle-income families ($P < 0.05$). However, the Ph concentrations in urine and in liquid food were higher for the middle-income children ($P < 0.05$). The PCP concentration in urine was higher for middle-income children ($P < 0.01$).

Table 2. Concentrations of the target compounds in indoor and outdoor air, floor dust and play area soil at two child day care centers.

Compound	Indoor Air (ng/m ³)		Outdoor air (ng/m ³)		Floor dust, ppm (μg/g)		Play area soil, ppm (μg/g)	
	Mean ^a	Range (N=4)	Mean ^a	Range (N=4)	Mean ^b	Range (N=4)	Mean ^b	Range (N=4)
Naphthalene	564	141–1044	55.6	39.4–76.5	0.027	0.008–0.046	0.001	<0.002–0.001 (2/4) ^c
Biphenyl	69.3	40.9–101	4.68	2.59–7.31	0.006	0.004–0.008	ND ^d	<0.002
Acenaphthylene	2.96	2.33–4.49	0.825	0.559–1.36	0.007	0.003–0.011	ND	<0.002
Acenaphthene	26.0	3.89–52.1	2.63	1.66–3.57	0.042	0.005–0.084	ND	<0.002
Fluorene	6.09	4.75–8.26	2.48	1.80–3.17	0.025	0.010–0.042	0.001	0.001
Phenanthrene	17.4	6.61–31.3	6.75	5.73–7.78	0.338	0.038–0.703	0.005	0.001–0.008
Anthracene	0.68	0.517–1.04	0.354	0.310–0.404	0.045	0.005–0.092	0.002	0.001–0.002
Fluoranthene	0.710	0.421–1.25	0.560	0.375–0.711	0.437	0.049–0.908	0.009	0.003–0.014
Pyrene	0.364	0.216–0.645	0.282	0.206–0.390	0.354	0.042–0.721	0.007	0.002–0.012
Cyclopenta[c,d]pyrene	ND	<0.040	ND	<0.040	0.055	0.010–0.110	0.002	0.001–0.003
Benz[a]anthracene* ^e	0.071	0.064–0.078	0.074	0.064–0.090	0.166	0.022–0.345	0.003	0.001–0.005
Chrysene*	0.099	0.082–0.128	0.070	0.056–0.092	0.213	0.046–0.424	0.004	0.001–0.007
Benzo[b]fluoranthene*	0.133	0.121–0.148	0.138	0.121–0.163	0.298	0.043–0.609	0.006	0.002–0.010
Benzo[k]fluoranthene*	0.085	0.068–0.092	0.095	0.088–0.106	0.113	0.016–0.236	0.003	0.001–0.005
Benzo[e]pyrene	0.058	0.051–0.066	0.061	0.052–0.074	0.159	0.027–0.319	0.003	0.001–0.005
Benzo[a]pyrene*	0.071	0.061–0.082	0.076	0.060–0.100	0.191	0.023–0.403	0.004	0.001–0.006
Indeno[1,2,3-c,d]pyrene*	0.097	0.088–0.107	0.102	0.091–0.115	0.197	0.032–0.396	0.004	0.002–0.006
Dibenz[a,h]anthracene*	0.055	0.048–0.070	ND	<0.040	0.073	0.015–0.144	0.003	0.002–0.004
Benzo[g,h,i]perylene	0.090	0.064–0.153	0.078	0.065–0.095	0.188	0.036–0.368	0.004	0.002–0.005
Coronene	0.088	0.073–0.122	0.087	0.077–0.093	0.059	0.017–0.104	0.002	0.001–0.002
Dibutyl phthalate	488	222–786	73.9	23.7–110	1.87	0.058–5.85	0.053	0.008–0.097
Benzylbutyl phthalate	144	128–151	133	102–192	3.72	0.022–7.43	0.032	<0.002–0.064 (2/4)
Diazinon	4.30	1.12–7.39	0.280	<0.100–0.570 (2/4)	0.034	0.011–0.066	0.001	<0.002–0.002 (2/4)
Chlorpyrifos	13.7	1.97–28.7	1.02	0.790–1.77	0.107	0.032–0.271	ND	<0.002
Lindane	5.74	4.38–6.81	0.340	0.260–0.490	0.019	0.018–0.020	0.005	0.004–0.005
Heptachlor	4.92	0.960–9.32	0.625	0.350–1.160	0.024	<0.002–0.048(2/4)	0.001	<0.002–0.002 (2/4)
Aldrin	2.08	<0.100–4.53 (2/4)	ND	<0.100	ND	<0.002	ND	<0.002
Gamma-chlordane	10.5	0.390–22.4	0.668	0.180–1.53	0.149	0.008–0.293	ND	<0.002
Alpha-chlordane	8.22	0.260–17.5	0.448	0.160–0.890	0.132	0.006–0.263	ND	<0.002
<i>p,p'</i> -DDE	0.115	<0.100–0.200 (2/4)	ND	<0.100	0.005	<0.002–0.013 (2/4)	ND	<0.002
Dieldrin	0.213	<0.100–0.580 (2/4)	ND	<0.100	0.009	<0.002–0.024 (2/4)	ND	<0.002
Endrin	0.333	0.300–0.370	ND	<0.100	0.070	<0.002–0.278 (3/4)	ND	<0.002
<i>p,p'</i> -DDT	ND	<0.100	ND	<0.100	0.011	<0.002–0.041 (3/4)	ND	<0.002
2,6 PCB	10.3	3.81–18.4	0.689	0.176–1.23	ND	<0.002	ND	<0.002
4,4' PCB	ND	<0.040	ND	<0.100	ND	<0.002	ND	<0.002
2,4,4' PCB	12.9	8.19–17.9	1.50	1.27–2.07	0.037	0.020–0.079	ND	<0.002
2,2',5,5' PCB	5.26	3.53–6.16	0.833	0.670–1.06	0.032	0.013–0.061	ND	<0.002
2,2',3,5' PCB	2.72	1.10–4.82	0.341	0.263–0.454	0.019	0.007–0.045	ND	<0.002
2,3',4',5' PCB	1.46	0.346–2.79	0.234	0.149–0.352	0.023	0.007–0.059	ND	<0.002
3,3',4,4' PCB	0.235	<0.040–0.881 (3/4)	ND	<0.100	0.004	<0.002–0.015 (3/4)	ND	<0.002
2,2',3,5',6 PCB	0.627	0.338–0.859	0.311	0.102–0.538	0.010	<0.002–0.026 (1/4)	ND	<0.002
2,2',4,5,5' PCB	0.647	0.376–0.862	0.391	0.192–0.748	0.019	0.007–0.045	ND	<0.002
2,2',3,4,5' PCB	0.312	0.197–0.456	0.189	0.115–0.345	0.014	0.008–0.030	ND	<0.002
2,3,3',4',6 PCB	0.502	0.221–0.839	0.314	0.114–0.598	0.028	0.012–0.069	ND	<0.002
2,3',4,4',5 PCB	0.301	0.078–0.612	0.156	0.040–0.267	0.020	0.010–0.047	ND	<0.002
2,3,3',4,4' PCB	0.220	0.138–0.364	0.133	0.093–0.180	0.011	<0.002–0.027 (1/4)	ND	<0.002
3,3',4,4',5 PCB	0.048	<0.040–0.091 (2/4)	0.029	<0.040–0.055(3/4)	ND	<0.002	ND	<0.002
2,2',4,4',5,5' PCB	0.219	0.105–0.424	0.090	<0.040–0.146 (1/4)	0.017	0.009–0.034	0.001	<0.002–0.001 (2/4)
2,2',3,4,4',5' PCB	0.222	0.113–0.495	0.138	0.044–0.288	0.019	0.007–0.044	0.001	0.001
3,3',4,4',5,5' PCB	ND	<0.040	ND	<0.100	0.007	<0.002–0.014 (2/4)	ND	<0.002
2,2',3,4,4',5,5' PCB	0.047	<0.040–0.077 (2/4)	0.026	<0.040–0.044 (3/4)	0.011	0.007–0.019	0.001	0.001–0.003
Pentachlorophenol	0.918	0.740–1.18	0.480	0.220–0.790	0.050	0.039–0.063	ND	<0.002
Nonyl phenols	253	165–392	2.76	0.060–5.47	29.2	4.62–52.6	0.063	0.059–0.070
Bisphenol-A	6.38	2.81–8.80	2.53	0.160–4.72	1.95	0.567–3.26	0.006	0.005–0.007
2,4-D	0.034	<0.040–0.074 (3/4)	0.058	<0.040–0.170	0.144	0.051–0.269	ND	<0.002
3,5,6-trichloro-2-pyridinol	NA ^f	NA	NA	NA	0.371	0.121–1.00	NA	NA

^aTwo air samples averaged over 48 h at each of the two day care centers.^bTwo dust and two soil samples collected at each day care center.^cThe detection limit is shown for those samples below the detection limit and the fractions of samples below the limit of detection are shown in parentheses. For those samples below the limit of detection, one-half the detection limit is used in the calculation of the mean.^dND indicates that all samples were below the limit of detection.^eAn asterisk indicates a B2 PAH.^fNA indicates that the samples were not analyzed for this analyte.

Table 3. Concentrations of the target compounds in indoor and outdoor air, floor dust and play area soil at homes of nine children.

Compound	Indoor air (ng/m ³)		Outdoor air (ng/m ³)		Floor dust ppm (μg/g)		Play area soil ppm (μg/g)	
	Mean ^a	Range (N=9)	Mean ^a	Range (N=9)	Mean	Range (N=9)	Mean	Range (N=9)
Naphthalene	413	153–1236	77.9	27.2–155	0.010	<0.002–0.035 (1/9) ^b	0.011	<0.002–0.095 (7/9)
Biphenyl	47.8	16.9–85.4	3.60	1.78–5.80	0.002	<0.002–0.005 (2/9)	ND ^c	<0.002
Acenaphthylene	3.48	1.72–5.40	0.780	0.308–1.63	0.006	0.002–0.023	0.001	<0.002–0.001 (8/9)
Acenaphthene	4.87	0.288–11.5	2.05	0.886–4.02	0.008	0.004–0.019	ND	<0.002
Fluorene	5.32	3.71–9.83	1.80	0.989–3.07	0.011	0.005–0.028	0.001	<0.002–0.004 (2/9)
Phenanthrene	9.18	5.38–19.0	3.52	1.76–7.85	0.144	0.044–0.596	0.008	<0.002–0.051 (2/9)
Anthracene	0.647	0.452–1.15	0.274	0.210–0.410	0.017	0.006–0.066	0.002	0.001–0.007
Fluoranthene	0.469	0.169–1.10	0.361	0.228–0.730	0.297	0.074–1.56	0.014	0.001–0.091
Pyrene	0.303	0.155–0.599	0.185	0.055–0.408	0.229	0.057–1.20	0.011	0.001–0.067
Cyclopenta[c,d]pyrene	0.038	<0.040–0.102(7/9)	ND	<0.040	0.035	0.010–0.172	0.003	<0.002–0.012 (1/9)
Benz[a]anthracene* ^d	0.076	0.061–0.109	0.067	0.060–0.092	0.090	0.019–0.519	0.005	<0.002–0.035 (4/9)
Chrysene*	0.108	0.076–0.172	0.064	0.054–0.098	0.169	0.037–0.838	0.007	0.001–0.041
Benzo[b]fluoranthene*	0.143	0.114–0.204	0.132	0.117–0.174	0.253	0.053–1.44	0.011	0.001–0.062
Benzo[k]fluoranthene*	0.095	0.089–0.111	0.090	0.086–0.109	0.050	0.019–0.496	0.004	<0.002–0.024 (2/9)
Benzo[e]pyrene	0.064	0.050–0.094	0.059	0.050–0.078	0.144	0.031–0.809	0.008	<0.002–0.033 (2/9)
Benzo[a]pyrene*	0.090	0.057–0.184	0.054	<0.040–0.105 (3/9)	0.134	0.024–0.768	0.007	<0.002–0.033
Indeno[1,2,3-c,d]pyrene*	0.110	0.086–0.149	0.056	0.087–0.116	0.169	0.035–0.963	0.007	<0.002–0.028 (1/9)
Dibenz[a,h]anthracene*	0.070	<0.040–0.129(1/9)	0.026	<0.040–0.071 (8/9)	0.060	0.016–0.294	0.004	<0.002–0.012 (1/9)
Benzo[g,h,i]perylene	0.085	0.064–0.111	0.073	0.063–0.092	0.173	0.040–0.961	0.010	<0.002–0.043 (1/9)
Coronene	0.089	0.077–0.107	0.077	<0.040–0.094 (1/9)	0.067	0.020–0.320	0.005	<0.002–0.024 (1/9)
Dibutyl phthalate	288	190–451	30.7	<1.00–84.7 (2/9)	1.21	0.384–3.03	0.092	<0.002–0.173(1/9)
Benzylbutyl phthalate	143	50.7–267	127	<1.00–474 (1/9)	5.86	0.496–15.6	0.039	<0.002–0.091 (1/9)
Diazinon	15.5	0.560–114	0.577	0.480–1.00	0.044	0.010–0.216	0.001	<0.002–0.002 (6/9)
Chlorpyrifos	158	2.90–1145	1.71	0.960–3.98	1.04	0.034–6.45	0.001	<0.002–0.009 (8/9)
Lindane	7.44	3.24–10.8	0.246	<0.100–0.590 (1/9)	0.033	0.014–0.046	0.005	0.003–0.006
Heptachlor	33.7	0.400–133	0.903	0.250–2.97	0.119	<0.002–0.335 (2/9)	0.002	<0.002–0.002 (1/9)
Aldrin	2.74	0.870–4.69	0.084	<0.100–0.250 (6/9)	0.006	<0.002–0.051 (8/9)	ND	<0.002
Gamma-chlordane	5.28	0.500–28.0	0.324	0.120–1.25	0.098	0.005–0.471	0.006	<0.002–0.048 (7/9)
Alpha-chlordane	2.45	0.320–13.0	0.248	0.120–0.740	0.055	0.004–0.256	0.005	<0.002–0.014 (7/9)
<i>p,p'</i> -DDE	0.124	<0.100–0.310(4/9)	0.059	<0.100–0.130 (8/9)	0.007	<0.002–0.047 (7/9)	0.004	<0.002–0.033 (8/9)
Dieldrin	0.184	<0.100–0.780(6/9)	0.056	<0.100–0.100 (8/9)	0.018	<0.002–0.050 (1/9)	ND	<0.002
Endrin	0.248	<0.100–0.690(5/9)	0.093	<0.100–0.440 (8/9)	ND	<0.002	ND	<0.002
<i>p,p'</i> -DDT	ND	<0.100	ND	<0.100	0.121	0.015–0.782	0.013	<0.002–0.110 (7/9)
2,6 PCB	2.55	<0.040–4.99	0.114	<0.040–0.326 (5/9)	ND	<0.002	ND	<0.002
4,4' PCB	ND	<0.040	ND	<0.040	ND	<0.002	ND	<0.002
2,4,4' PCB	1.93	0.783–3.79	0.321	0.172–0.468	0.005	<0.002–0.018 (5/9)	ND	<0.002
2,2',5,5' PCB	3.58	0.455–5.18	0.363	0.231–0.603	0.009	<0.002–0.026 (4/9)	ND	<0.002
2,2',3,5' PCB	6.69	0.624–51.4	0.105	<0.040–0.283 (2/9)	0.006	<0.002–0.013 (4/9)	ND	<0.002
2,3',4',5' PCB	0.387	0.190–0.530	0.096	<0.040–0.229 (3/9)	0.008	<0.002–0.017 (3/9)	ND	<0.002
3,3',4',5' PCB	0.067	<0.040–0.445 (8/9)	ND	<0.040	0.002	<0.002–0.006 (7/9)	ND	<0.002
2,2',3,5',6' PCB	0.256	<0.040–0.510 (2/9)	0.137	<0.040–0.287 (2/9)	0.004	<0.002–0.016 (6/9)	ND	<0.002
2,2',4,5,5' PCB	0.326	0.142–0.670	0.174	<0.040–0.426 (2/9)	0.014	<0.002–0.036 (2/9)	0.001	<0.002–0.002 (8/9)
2,2',3,4,5' PCB	0.168	0.046–0.358	0.078	<0.040–0.232 (4/9)	0.011	<0.002–0.024 (1/9)	ND	<0.002
2,3,3',4',6' PCB	0.254	0.039–0.714	0.116	<0.040–0.413 (4/9)	0.018	0.006–0.044	0.001	<0.002–0.003 (7/9)
2,3',4,4',5' PCB	0.186	0.057–0.547	0.091	<0.040–0.428 (4/9)	0.014	<0.002–0.035 (2/9)	0.001	<0.002–0.002 (7/9)
2,3,3',4,4' PCB	0.137	<0.040–0.267 (3/9)	0.111	<0.040–0.235 (1/9)	0.006	<0.002–0.018 (5/9)	ND	<0.002
3,3',4,4',5' PCB	0.023	<0.040–0.046 (8/9)	0.025	<0.040–0.066 (8/9)	ND	<0.002	0.001	<0.002–0.002 (8/9)
2,2',4,4',5,5' PCB	0.109	<0.040–0.327 (3/9)	0.057	<0.040–0.213 (5/9)	0.013	0.004–0.025	0.001	<0.002–0.003 (7/9)
2,2',3,4,4',5' PCB	0.142	<0.040–0.414 (2/9)	0.160	<0.040–0.415 (2/9)	0.012	0.005–0.022	0.001	<0.002–0.003 (7/9)
3,3',4,4',5,5' PCB	ND	<0.040	0.041	<0.040–0.108 (6/9)	0.010	<0.002–0.022 (2/9)	ND	<0.002
2,2',3,4,4',5,5' PCB	0.031	<0.040–0.117 (8/9)	0.025	<0.040–0.061 (8/9)	0.010	0.005–0.022	0.001	<0.002–0.004 (7/9)
Pentachlorophenol	9.11	0.660–53.2	0.244	0.100–0.600	0.135	0.034–0.571	ND	<0.002
Nonyl phenols	169	0.310–402	2.42	1.06–4.36	7.22	3.28–9.62	0.076	0.034–0.162
Bisphenol-A	11.8	<0.100–29.0 (1/9)	1.26	<0.100–4.41 (4/9)	1.52	0.707–1.89	0.007	0.004–0.014
2,4-D	0.085	<0.040–0.313 (7/9)	0.025	<0.040–0.069 (8/9)	1.24	0.0247–7.29	0.032	<0.002–0.151 (2/9)
3,5,6-trichloro-2-pyridinol	NA ^e	NA	NA	NA	0.535 ^f	0.206–0.950	NA	NA

^aOne air sample averaged over 48 h at each of the nine homes.

^bThe detection limit is shown for those samples below the detection limit and the fractions of samples below the limit of detection are shown in parentheses. For those samples below the limit of detection, one-half the detection limit is used in the calculation of the mean.

^cND indicates that all samples were below the limit of detection.

^dAn asterisk indicates a B2 PAH.

^eNA indicates that the samples were not analyzed for this analyte.

^fSamples from five homes.

Table 4. Concentrations of the target compounds in liquid food, solid food, and hand wipes, for nine children at day care.

Compound	Liquid Food, ppb (ng/g)		Solid Food, ppb (ng/g)		Hand Wipes, ng/wipe	
	Mean ^a	Range (N=4)	Mean ^a	Range (N=4)	Mean ^b	Range (N=9)
Naphthalene	0.170	<0.040–0.547 (1/4) ^c	2.04	0.635–2.72	2.01	0.860–2.70
Biphenyl	0.026	<0.040–0.042 (3/4)	0.202	<0.040–0.360 (1/4)	1.20	0.470–1.71
Acenaphthylene	0.104	<0.040–0.144 (1/4)	0.377	<0.040–1.20 (2/4)	ND ^d	<0.500
Acenaphthene	0.360	<0.040–1.381 (3/4)	ND	<0.040	3.66	0.895–6.22
Fluorene	0.114	0.097–0.126	0.583	0.200–1.13	2.02	<0.500–5.30 (3/9)
Phenanthrene	0.036	<0.040–0.058 (1/4)	0.510	0.176–0.914	1.382	<0.500–5.45 (4/9)
Anthracene	ND	<0.040	ND	<0.040	0.596	<0.500–1.90(3/9)
Fluoranthene	0.043	<0.040–0.059 (1/4)	0.262	0.129–0.467	1.15	<0.500–8.20 (7/9)
Pyrene	0.055	<0.040–0.091 (2/4)	0.164	0.042–0.280	0.774	<0.500–4.56 (6/9)
Cyclopenta[c,d]pyrene	ND	<0.040	ND	<0.040	0.274	<0.500–0.465 (8/9)
Benz[a]anthracene* ^c	0.126	<0.040–0.349 (2/4)	ND	<0.040	0.315	<0.500–0.835 (8/9)
Chrysene*	0.040	<0.040–0.101 (3/4)	ND	<0.040	0.376	<0.500–1.02 (7/9)
Benzo[b]fluoranthene*	ND	<0.040	ND	<0.040	0.383	<0.500–1.45(8/9)
Benzo[k]fluoranthene*	ND	<0.040	ND	<0.040	0.274	<0.500–0.465 (8/9)
Benzo[e]pyrene	ND	<0.040	ND	<0.040	0.308	<0.500–0.770 (8/9)
Benzo[a]pyrene*	ND	<0.040	ND	<0.040	0.388	<0.500–1.29 (7/9)
Indeno-[1,2,3-c,d]pyrene*	ND	<0.040	ND	<0.040	0.532	<0.500–1.24 (3/9)
Dibenz[a,h]anthracene*	ND	<0.040	ND	<0.040	0.304	<0.500–0.735 (8/9)
Benzo[g,h,i]perylene	ND	<0.040	ND	<0.040	0.586	<0.500–1.21 (1/9)
Coronene	ND	<0.040	ND	<0.040	ND	<0.500
Dibutyl phthalate	6.10	<0.040–23.4 (2/4)	35.6	5.39–73.0	97.0	<0.500–605 (4/9)
Benzylbutyl phthalate	6.54	4.25–7.57	31.4	8.68–53.7	306	<0.500–1325 (3/9)
Diazinon	ND	<0.040	ND	<0.040	1.30	<0.500–9.33 (7/9)
Chlorpyrifos	0.094	<0.040–0.317 (3/4)	0.651	0.214–1.65	1.56	<0.500–5.26 (5/9)
Lindane	ND	<0.040	ND	<0.040	ND	<0.500
Heptachlor	0.367	<0.040–0.557 (1/4)	ND	<0.040	ND	<0.500
Aldrin	ND	<0.040	ND	<0.040	ND	<0.500
Gamma-chlordane	0.041	<0.040–0.069 (2/4)	ND	<0.040	0.724	<0.500–3.05 (5/9)
Alpha-chlordane	0.027	<0.040–0.049 (3/4)	ND	<0.040	0.521	<0.500–2.06 (6/9)
p,p'-DDE	0.029	<0.040–0.054 (3/4)	0.067	<0.040–0.138 (2/4)	ND	<0.500
Dieldrin	ND	<0.040	ND	<0.040	ND	<0.500
Endrin	ND	<0.040	ND	<0.040	ND	<0.500
p,p'-DDT	0.114	<0.040–0.221 (2/4)	0.103	<0.040–0.350 (3/4)	0.383	<0.500–1.45 (8/9)
2,6 PCB	0.034	<0.040–0.077 (3/4)	ND	<0.040	ND	<0.500
4,4' PCB	0.045	<0.040–0.120 (3/4)	ND	<0.040	ND	<0.500
2,4,4' PCB	ND	<0.040	ND	<0.040	ND	<0.500
2,2',5,5' PCB	ND	<0.040	0.045	<0.040–0.119 (3/4)	0.297	<0.500–0.670 (8/9)
2,2',3,5' PCB	ND	<0.040	ND	<0.040	0.372	<0.500–1.01 (7/9)
2,3',4',5' PCB	ND	<0.040	ND	<0.040	ND	<0.500
3,3',4,4' PCB	ND	<0.040	ND	<0.040	ND	<0.500
2,2',3,5',6' PCB	ND	<0.040	0.041	<0.040–0.102	ND	<0.500
2,2',4,5,5' PCB	ND	<0.040	ND	<0.040	ND	<0.500
2,2',3,4,5' PCB	ND	<0.040	ND	<0.040	ND	<0.500
2,3,3',4',6' PCB	0.045	<0.040–0.119 (3/4)	ND	<0.040	0.352	<0.500–0.890 (7/9)
2,3',4,4',5' PCB	ND	<0.040	ND	<0.040	0.493	<0.500–0.920 (5/9)
2,3,3',4,4' PCB	ND	<0.040	ND	<0.040	ND	<0.500
3,3',4,4',5' PCB	ND	<0.040	ND	<0.040	ND	<0.500
2,2',4,4',5,5' PCB	0.029	<0.040–0.054 (3/4)	ND	<0.040	ND	<0.500
2,2',3,4,4',5' PCB	ND	<0.040	ND	<0.040	ND	<0.500
3,3',4,4',5,5' PCB	ND	<0.040	ND	<0.040	ND	<0.500
2,2',3,4,4',5,5' PCB	ND	<0.040	ND	<0.040	ND	<0.500
Pentachlorophenol	ND	<0.040	ND	<0.100	NA ^f	NA
Nonyl phenols	2.57	<0.100–4.32 (1/4)	19.9	10.5–34.2	NA	NA
Bisphenol-A	0.384	<0.100–1.16 (1/4)	0.513	0.196–0.932	NA	NA
2,4-D	1.59	0.917–2.01	0.717	<0.500–1.45 (2/4)	NA	NA
3,5,6-trichloro-2-pyridinol	1.0	0.39–2.0	16	14–18	NA	NA

^aAll children were served the same foods at day care. Two daily composite samples were obtained at each of the two child day care centers.

^bComposite of four hand-wipe samples over two days at day care for each of nine children.

^cThe detection limit is shown for those samples below the detection limit and the fractions of samples below the limit of detection are shown in parentheses. For those samples below the limit of detection, one-half the detection limit is used in the calculation of the mean.

^dND indicates that all samples were below the limit of detection.

^eAn asterisk indicates a B2 PAH.

^fNA indicates not analyzed.

Table 5. Concentrations of the target compounds in liquid food, solid food, and hand wipes, for nine children at home.

Compound	Liquid food, ppb (ng/g)		Solid food, ppb (ng/g)		Hand wipes, ng/wipe	
	Mean ^a	Range (N=9)	Mean ^a	Range (N=9)	Mean ^b	Range (N=9)
Naphthalene	0.023	<0.040–0.043 (8/9) ^c	1.83	0.782–2.77	3.90	<0.500–22.5 (2/9)
Biphenyl	ND ^d	<0.040	0.327	<0.040–1.03 (2/9)	0.844	<0.500–1.70 (3/9)
Acenaphthylene	0.070	<0.040–0.133 (4/9)	0.196	<0.040–1.23 (4/9)	ND	<0.500
Acenaphthene	ND	<0.040	ND	<0.040	3.24	<0.500–6.88 (1/9)
Fluorene	0.074	<0.040–0.139 (3/9)	0.498	0.230–0.814	1.15	<0.500–2.23 (3/9)
Phenanthrene	0.066	<0.040–0.345 (7/9)	0.594	0.273–1.10	0.867	<0.500–4.73 (6/9)
Anthracene	ND	<0.040	ND	<0.040	0.392	<0.500–0.695 (5/9)
Fluoranthene	0.042	<0.040–0.185 (7/9)	0.259	0.158–0.427	0.301	<0.500–0.690 (7/9)
Pyrene	0.070	<0.040–0.139 (1/9)	0.160	0.085–0.287	0.308	<0.500–0.550 (7/9)
Cyclopenta[c,d]pyrene	ND	<0.040	ND	<0.040	ND	<0.500
Benz[a]anthracene* ^c	0.031	<0.040–0.120 (8/9)	ND	<0.040	ND	<0.500
Chrysene*	0.029	<0.040–0.098 (8/9)	ND	<0.040	0.272	<0.500–0.450 (8/9)
Benzo[b]fluoranthene*	ND	<0.040	ND	<0.040	0.285	<0.500–0.565 (8/9)
Benzo[k]fluoranthene*	ND	<0.040	ND	<0.040	0.304	<0.500–0.735 (8/9)
Benzo[e]pyrene	ND	<0.040	ND	<0.040	ND	<0.500
Benzo[a]pyrene*	ND	<0.040	ND	<0.040	0.359	<0.500–0.610 (6/9)
Indeno[1,2,3-c,d]pyrene*	ND	<0.040	ND	<0.040	0.492	<0.500–0.895 (4/9)
Dibenz[a,h]anthracene*	ND	<0.040	ND	<0.040	0.317	<0.500–0.595 (7/9)
Benzo[g,h,i]perylene	ND	<0.040	ND	<0.040	0.566	<0.500–0.780 (1/9)
Coronene	ND	<0.040	ND	<0.040	ND	<0.500
Dibutyl phthalate	10.4	<0.040–55.6 (4/9)	20.5	2.75–57.4	72.1	<0.500–184 (4/9)
Benzylbutyl phthalate	9.20	2.68–15.1	100	8.59–404	297	<0.500–938 (2/9)
Diazinon	0.064	<0.040–0.234 (7/9)	ND	<0.040	1.17	<0.500–8.06 (7/9)
Chlorpyrifos	0.066	<0.040–0.432 (8/9)	0.819	0.085–2.31	5.78	<0.500–23.9 (2/9)
Lindane	ND	<0.040	ND	<0.040	ND	<0.500
Heptachlor	0.104	<0.040–0.421 (7/9)	0.304	<0.040–0.818 (5/9)	ND	<0.500
Aldrin	ND	<0.040	ND	<0.040	ND	<0.500
Gamma-chlordane	0.035	<0.040–0.159 (8/9)	0.033	<0.040–0.135 (8/9)	1.17	<0.500–6.45 (5/9)
Alpha-chlordane	0.025	<0.040–0.066 (8/9)	0.025	<0.040–0.063 (8/9)	0.905	<0.500–4.05 (6/9)
<i>p,p'</i> -DDE	0.033	<0.040–0.099 (7/9)	0.163	<0.040–1.31 (8/9)	ND	<0.500
Dieldrin	0.020	<0.040–0.024 (8/9)	ND	<0.040	ND	<0.500
Endrin	ND	<0.040	ND	<0.040	ND	<0.500
<i>p,p'</i> -DDT	0.142	<0.040–0.243 (3/9)	0.115	<0.040–0.769 (6/9)	0.311	<0.500–0.800 (8/9)
2,6 PCB	ND	<0.040	ND	<0.040	ND	<0.500
4,4' PCB	0.047	<0.040–0.134 (6/9)	0.045	<0.040–0.175 (6/9)	ND	<0.500
2,4,4' PCB	ND	<0.040	0.053	<0.040–0.109 (5/9)	ND	<0.500
2,2',5,5' PCB	ND	<0.040	0.034	<0.040–0.142 (8/9)	ND	<0.500
2,2',3,5' PCB	ND	<0.040	0.026	<0.040–0.072 (8/9)	0.329	<0.500–0.965 (8/9)
2,3',4',5' PCB	ND	<0.040	0.029	<0.040–0.102 (8/9)	0.281	<0.500–525 (8/9)
3,3',4,4' PCB	ND	<0.040	0.032	<0.040–0.088 (7/9)	ND	<0.500
2,2',3,5',6' PCB	ND	<0.040	0.027	<0.040–0.087 (8/9)	ND	<0.500
2,2',4,5,5' PCB	ND	<0.040	0.030	<0.040–0.112 (8/9)	ND	<0.500
2,2',3,4,5' PCB	ND	<0.040	0.023	<0.040–0.047 (8/9)	ND	<0.500
2,3,3',4',6' PCB	ND	<0.040	0.033	<0.040–0.134 (8/9)	0.316	<0.500–0.560 (7/9)
2,3',4,4',5' PCB	ND	<0.040	0.025	0.040–0.061 (8/9)	0.408	<0.500–0.955 (6/9)
2,3,3',4,4' PCB	ND	<0.040	ND	<0.040	ND	<0.500
3,3',4,4',5' PCB	ND	<0.040	ND	<0.040	ND	<0.500
2,2',4,4',5,5' PCB	ND	<0.040	0.029	<0.040–0.105 (8/9)	ND	<0.500
2,2',3,4,4',5' PCB	ND	<0.040	0.027	<0.040–0.082 (8/9)	ND	<0.500
3,3',4,4',5,5' PCB	ND	<0.040	ND	<0.040	ND	<0.500
2,2',3,4,4',5,5' PCB	ND	<0.040	ND	<0.040	ND	<0.500
Pentachlorophenol	ND	<0.100	0.063	<0.100–0.116 (7/9)	NA ^f	NA
Nonyl phenols	0.833	<0.100–3.28 (5/9)	32.6	12.5–76.1	NA	NA
Bisphenol-A	0.069	<0.100–0.145 (7/9)	1.30	0.172–4.19	NA	NA
2,4-D	1.46	0.353–3.19	0.45	<0.500–2.20 (8/9)	NA	NA
3,5,6-trichloro-2-pyridinol	0.91	0.095–3.6	20	6.5–43	NA	NA

^aComposite of two daily samples at home for each child.

^bComposite of two hand-wipe samples over 2 days at home for each child.

^cThe detection limit is shown for those samples below the detection limit and the fractions of samples below the limit of detection are shown in parentheses. For those samples below the limit of detection, one-half the detection limit is used in the calculation of the mean.

^dND indicates that all samples were below the limit of detection.

^eAn asterisk indicates a B2 PAH.

^fNA indicates not analyzed.

Table 6. Concentrations of targeted compounds in composited urine samples over 48 h for nine children at home and day care^a.

Compound	Mean (ng/ml)	Range (ng/ml) (N=9)	Mean (μ mol/mol) ^b	Range (μ mol/mol) (N=9)
Pentachlorophenol	0.329	0.175–0.666	0.458	0.113–1.11
2,4-D	2.26	0.710–3.49	2.86	1.79–4.08
1-Naphthol	0.585	0.231–1.47	1.37	0.260–3.03
2-Naphthol	0.156	0.087–0.272	0.356	0.134–0.548
3-Hydroxyfluoranthene	0.166	0.064–0.397	0.288	0.043–0.895
1-Hydroxypyrene	0.078	0.021–0.155	0.127	0.029–0.315
1-Hydroxybenz[a]anthracene	0.023	<0.018–0.043 (3/9) ^c	0.035	<0.020–0.105 (3/9)
6-Hydroxychrysene	0.050	0.020–0.150	0.083	0.026–0.289
3-Hydroxybenz[a]anthracene	0.044	<0.018–0.079 (1/9)	0.059	<0.033–0.133 (1/9)
1- and 3-Hydroxybenzo[a]pyrene	0.024	<0.033–0.061 (7/9)	0.033	<0.018–0.107 (7/9)
6-Hydroxyindeno[1,2,3-c,d]pyrene	0.023	<0.018–0.076 (4/9)	0.023	<0.016–0.049 (4/9)
3,5,6-Trichloro-2-pyridinol	8.52	3.76–17.7	14.9	4.99–39.2

^aComposites of four samples at home (first morning and before bedtime) and two at day care (after lunch) for each child over 2 days.

^bNormalized to micromoles per mole of creatinine.

^cThe detection limit is shown for those samples below the detection limit and the fractions of samples below the limit of detection are shown in parentheses. For those samples below the limit of detection, one-half the detection limit is used in the calculation of the mean.

The differences between day care and home, and between low- and middle-income, while statistically significant, are small, and do not give a clear-cut answer to the questions posed in the second and third hypotheses above. Examination of both effects, day care vs. home location and low- vs. middle-income, requires additional research to obtain data for a larger number of participants.

Potential Doses

The aggregate daily potential doses (PDs) of the targeted compounds through the inhalation, dietary ingestion, and nondietary ingestion pathways for the individual children are shown in Table 7, and illustrated in Figure 1. In the last four columns of the table are the mean, median, and range of potential doses. The volatile, abundant PAH with two or three rings contribute substantially to the total PAH exposure and PD. However, the seven PAH that are probable carcinogens together contributed a mean PD of about 10 ng/kg per day to the children in this study, with a range of about 2–33 and a median PD of 5 ng/kg day. The PE total PD values shown are probably underestimates and do not reflect the overall phthalate exposures, since many phthalates that were not measured in this study, such as diethylhexyl phthalate, are in high abundance in the environment. The two organophosphate pesticides, CPF and diazinon, were widely used at the time of this study (summer 1997), and the PDs for these two OPs reflect this use, especially the use of CPF in residences and schools. Many of the organochlorine pesticides that are no longer in use have a long lifetime in the environment and still contribute to the PD for the children studied. Of the Ph, the nonyl phenols contributed most to the PD for this class of compounds. The herbicide 2,4-D is in wide use on lawns and play areas, and all nine children had

exposures to this compound, with PDs ranging from 17 to 149 ng/kg day.

The PDs through inhalation and ingestion for each child were compared to the published daily oral reference doses (RfDs) (IRIS), for those chemicals for which RfD values are available. For most of the compounds, the mean ratio RfD/PD ranged from 44 to 410 000, and the maximum ratio ranged from 20 to 36 100, indicating that the pollutant concentrations were well within safe levels. Surprisingly, two OC pesticides that are no longer in use had mean RfD/PDs of 14 and 16, and at the maximum PDs, RfD/PDs of 7 and 6, for aldrin and heptachlor, respectively. The PDs of the two OP pesticides were closest to the RfD values: CPF with a mean RfD/PD of 27 and at the maximum PD, an RfD/PD of 5; and diazinon with a mean RfD/PD of 8 and at the maximum PD, an RfD/PD of 1. The latter maximum PD value was obtained for a child whose household had a pesticide application approximately 3 days before we collected our samples. Thus, the child may have exceeded the RfD for several days. In future studies, it would be desirable to collect detailed information on pesticide applications and the timing of these applications relative to the sampling period, so that interpretation of the results and understanding of the significance of these occasional higher levels are less difficult.

Routes of Exposure

Figure 2 exhibits the children's mean daily potential doses, in ng/kg day, of the targeted compound classes through the inhalation, dietary ingestion, and nondietary ingestion routes of exposure. It can be seen that the highest exposures found in this study are to the PE, the Ph, and TCP, all of which are associated primarily with dietary ingestion. The mean potential dose of TCP from nondietary ingestion was

Table 7. Estimated aggregate daily doses of the target compounds from homes and day care centers for nine children.

Compound	Potential dose (ng/kg day)												
	Participant									Mean	Maximum	Minimum	Median
	HA3	HB3	HC3	HD3	HE9	HF9	HG9	HH9	HI9				
Naphthalene	283.893	313.854	392.801	175.690	213.503	678.719	161.688	99.587	187.489	278.580	678.719	99.587	213.503
Biphenyl	44.108	43.031	32.282	17.494	35.260	35.501	31.711	19.518	23.279	31.354	44.108	17.494	32.282
Acenaphthylene	12.387	6.933	21.847	13.904	5.369	11.879	9.392	7.754	9.148	10.957	21.847	5.369	9.392
Acenaphthene	42.154	43.489	6.266	6.841	2.150	4.144	2.132	2.066	0.969	12.246	43.489	0.969	4.144
Fluorene	27.299	30.617	16.389	8.110	17.831	21.490	24.704	22.940	27.374	21.861	30.617	8.110	22.940
Phenanthrene	21.222	24.173	26.573	15.278	26.053	36.240	28.403	13.786	24.143	23.986	36.240	13.786	24.173
Anthracene	0.706	0.557	0.532	0.332	0.318	0.785	0.282	0.371	0.340	0.469	0.785	0.282	0.371
Fluoranthene	8.137	10.536	12.128	6.736	10.966	21.855	12.261	5.276	11.240	11.015	21.855	5.276	10.966
Pyrene	6.350	9.692	8.983	4.966	7.234	16.559	8.574	5.796	7.729	8.431	16.559	4.966	7.729
Cyclopenta[c,d]pyrene	0.230	0.320	0.269	0.144	0.069	0.912	0.090	0.092	0.137	0.251	0.912	0.069	0.144
Benz[a]anthracene	8.643	10.578	0.711	0.444	3.628	7.271	0.217	0.199	0.238	3.548	10.578	0.199	0.711
Chrysene	3.286	4.644	0.957	0.603	0.324	4.485	0.757	0.398	0.519	1.775	4.644	0.324	0.757
Benzo[b]fluoranthene	1.350	1.767	1.428	0.834	0.490	7.642	0.573	0.629	0.652	1.707	7.642	0.490	0.834
Benzo[k]fluoranthene	0.530	0.700	0.545	0.336	0.210	2.675	0.228	0.251	0.269	0.638	2.675	0.210	0.336
Benzo[e]pyrene	0.720	0.930	0.751	0.448	0.278	4.296	0.329	0.367	0.381	0.944	4.296	0.278	0.448
Benzo[a]pyrene	0.847	1.123	0.836	0.506	0.271	4.086	0.312	0.331	0.350	0.963	4.086	0.271	0.506
Indeno[1,2,3-c,d]pyrene	0.879	1.158	0.869	0.559	0.353	5.130	0.401	0.446	0.478	1.141	5.130	0.353	0.559
Dibenz[a,h]anthracene	0.351	0.456	0.367	0.231	0.139	1.603	0.175	0.186	0.223	0.415	1.603	0.139	0.231
Benzo[g,h,i]perylene	0.846	1.101	0.833	0.552	0.361	5.120	0.418	0.441	0.500	1.130	5.120	0.361	0.552
Coronene	0.304	0.383	0.306	0.212	0.193	1.763	0.247	0.213	0.293	0.435	1.763	0.193	0.293
Dibutylphthalate	745.015	791.795	1307.165	899.936	2350.170	2853.106	1873.821	1137.262	989.036	1438.590	2853.106	745.015	1137.262
Benzylbutylphthalate	2053.118	1961.564	1326.346	744.161	2646.389	2764.006	1705.798	1063.473	2875.824	1904.520	2875.824	744.161	1961.564
Diazinon	42.973	4.754	1.639	3.001	0.586	1.732	0.731	0.981	1.439	6.426	42.973	0.586	1.639
Chlorpyrifos	30.006	19.313	10.622	16.198	56.561	151.550	21.936	329.318	49.946	76.161	329.318	10.622	30.006
Lindane	4.551	4.083	4.452	1.491	3.042	3.511	3.416	2.772	3.231	3.394	4.551	1.491	3.416
Heptachlor	22.660	6.052	17.839	6.608	13.927	21.568	54.084	58.769	16.930	24.271	58.769	6.052	17.839
Aldrin	2.024	2.222	2.312	0.735	0.494	1.891	0.209	0.391	0.575	1.206	2.312	0.209	0.735
Gamma-chlordane	4.130	5.205	4.049	3.820	2.039	5.515	8.180	3.783	0.618	4.149	8.180	0.618	4.049
Alpha-chlordane	3.184	4.095	2.408	2.349	1.805	3.722	3.912	1.461	0.346	2.587	4.095	0.346	2.408
p,p'-DDE	1.570	1.395	0.092	0.284	0.000	0.057	3.730	4.197	31.279	4.734	31.279	0.000	1.395
Dieldrin	0.383	0.149	0.142	0.167	0.026	0.586	0.013	0.058	0.173	0.189	0.586	0.013	0.149
Endrin	0.279	0.064	0.649	0.362	0.154	0.080	0.147	0.051	0.064	0.206	0.649	0.051	0.147
p,p'-DDT	10.043	6.042	6.467	4.069	8.363	13.890	0.110	7.427	18.225	8.293	18.225	0.110	7.427
Pentachlorophenol	1.449	4.153	0.867	0.417	1.568	1.676	3.226	15.559	3.264	3.575	15.559	0.417	1.676
Nonyl phenols	870.274	928.738	574.036	212.259	1344.552	1578.793	1523.322	927.052	969.975	992.111	1578.793	212.259	928.738
Bisphenol-A	43.833	66.583	43.212	20.644	44.523	53.359	71.124	18.466	25.087	42.981	71.124	18.466	43.833
2,4-D	87.609	60.530	52.936	17.174	114.261	149.009	75.081	128.789	93.615	86.556	149.009	17.174	87.609
Sum of all PAH	464.240	506.042	525.672	254.219	325.000	872.156	282.893	180.648	295.752	411.847	872.156	180.648	325.000
Sum of B2 PAH	15.886	20.426	5.714	3.512	5.415	32.893	2.663	2.441	2.730	10.187	32.893	2.441	5.415
Sum of phthalates	2798.133	2753.359	2633.512	1644.097	4996.559	5617.112	3579.619	2200.735	3864.860	3343.110	5617.112	1644.097	2798.133
Sum of OP pesticides	72.980	24.066	12.261	19.199	57.148	153.282	22.667	330.299	51.385	82.587	330.299	12.261	51.385
Sum of OC pesticides	48.824	29.308	38.411	19.883	29.850	50.821	73.802	78.908	71.442	49.028	78.908	19.883	48.824
Sum of PCBs	13.289	12.111	13.956	8.319	15.720	16.433	18.563	31.255	30.527	17.797	31.255	8.319	15.720
Sum of phenols	915.556	999.473	618.116	233.320	1390.643	1633.828	1597.671	961.078	998.326	1038.668	1633.828	233.320	998.326
	Potential dose (nmol/kg day)												
2,4-D	0.398	0.275	0.241	0.078	0.519	0.677	0.341	0.585	0.426	0.393	0.677	0.078	0.398
Sum of all PAH	3.294	3.580	3.849	1.836	2.318	6.344	1.973	1.256	2.095	2.949	6.344	1.256	2.318
Sum of B2 PAH	0.068	0.087	0.023	0.014	0.023	0.133	0.011	0.010	0.011	0.042	0.133	0.010	0.023
Sum of phthalates	9.260	9.135	8.953	5.622	16.936	19.122	12.208	7.499	12.775	11.279	19.122	5.622	9.260
Sum of OP pesticides	0.227	0.071	0.036	0.056	0.163	0.437	0.065	0.941	0.147	0.238	0.941	0.036	0.147
Sum of OC pesticides	0.135	0.081	0.106	0.054	0.083	0.138	0.199	0.215	0.211	0.136	0.215	0.054	0.135
Sum of PCBs	0.043	0.039	0.045	0.027	0.051	0.053	0.060	0.101	0.099	0.058	0.101	0.027	0.051
Sum of phenols	4.219	4.629	2.867	1.088	6.379	7.497	7.355	4.381	4.569	4.776	7.497	1.088	4.569

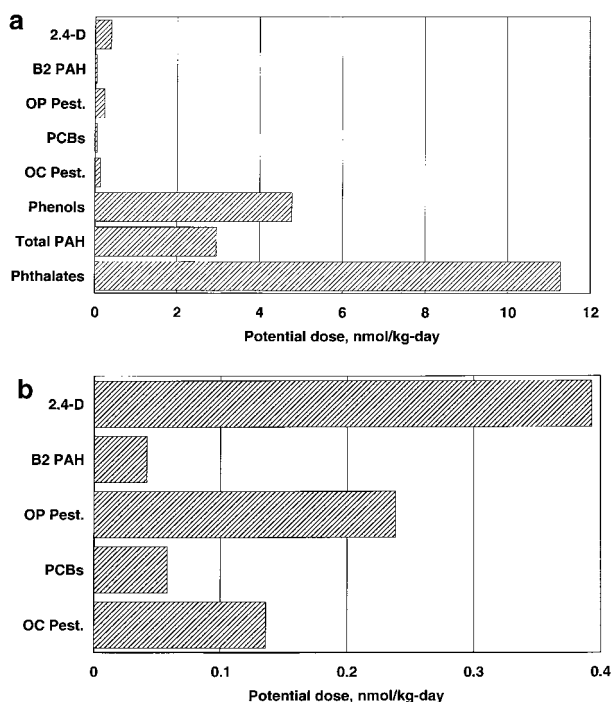


Figure 1. The calculated aggregate daily potential doses of the targeted groups of chemicals for nine children at home and day care, in nmol/kg day. (a) PDs for all measured groups of chemicals. (b) PDs with those of phthalates, Ph, and total PAH omitted.

3.06 ng/kg day. The air samples in this study were not analyzed for TCP, so the potential dose of this compound from inhalation was not calculated here. However, archived air samples from a previous study of children's exposures, which took place in the same geographic location (Chuang et al., 1999a), were analyzed. Three outdoor air samples had mean TCP concentrations of 0.3 ng/m³, and one indoor air sample had a TCP concentration of 1.7 ng/m³, which would produce an estimated mean potential dose of TCP of 0.6 ng/kg day from inhalation. Such estimated contributions of TCP from non-dietary ingestion and inhalation pale beside the potential dose from dietary ingestion measured here, which is 504 ng/kg day, as seen in Figure 2a.

The total PAH, which include both the known human carcinogens (B2 PAH), most of which are present at low concentrations in the environmental media, and the more abundant and more volatile PAH, such as naphthalene, anthracene, and phenanthrene, are associated primarily with inhalation and secondarily with dietary ingestion. On the expanded scale in Figure 2b, the role of dietary ingestion is clear for most of the compound classes, and is the primary route of exposure for 2,4-D. Inhalation and dietary ingestion are both important routes of exposure for the organophosphate and organochlorine pesticides and the PCBs.

In the NHEXAS-Arizona study, the concentrations of the OP pesticide CPF in air were well correlated with hand-wipe concentrations, although those in dust were not (Gordon

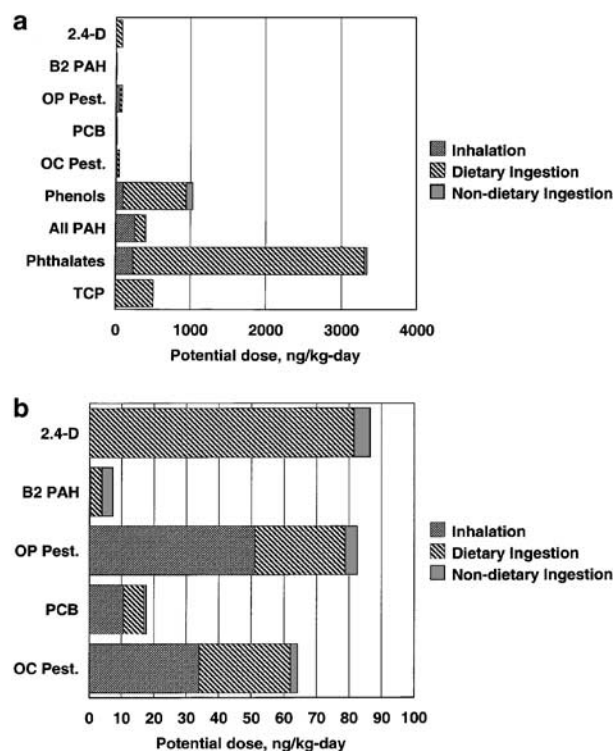


Figure 2. The calculated daily PDs of the targeted groups of chemicals for nine children at home and day care through the inhalation, dietary ingestion, and nondietary ingestion routes of exposure, in ng/kg day. Inhalation doses of TCP were not measured in this study. (a) PDs for all measured groups of chemicals. (b) PDs with those of phthalates, Ph, TCP, and total PAH omitted.

et al., 1999). This suggests that non dietary exposure to this class of compounds, through hand-to-mouth transfer, may contribute to the aggregate exposure. However, the contributions of nondietary exposure found in the study reported here are relatively small for most of the target compound classes, and appear to be significant, relative to other routes, mainly for the B2 PAH, most of which have five or more rings and are of very low volatility.

Biomarkers of Exposure

The children in this study in general had low, likely chronic, exposures to several of the target compounds. For two compounds measured in urine, 2,4-D and PCP, the urinary concentrations were correlated significantly with the children's aggregate daily potential doses. However, the urinary concentrations of the CPF metabolite TCP were not significantly correlated with the potential doses.

A steady-state scenario between exposure and excretion of urinary metabolites of the compounds or the unchanged compounds themselves was assumed, and the children's daily potential doses of PCP, 2,4-D, benz[a]anthracene (BaA), and CPF were compared to the excreted doses measured in their urine. The urinary concentrations are given in Table 6. Daily urine outputs of 22.4 ml/kg body weight for the

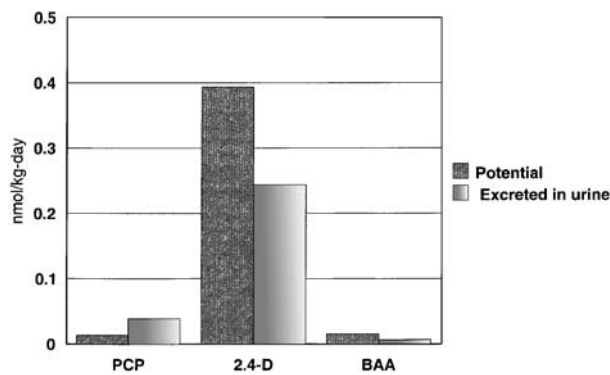


Figure 3. Mean aggregate daily potential and excreted doses of pentachlorophenol, 2,4-D, and benz[a]anthracene from ingestion and inhalation for nine children, in nmol/kg day.

children were assumed (Ballauf et al., 1988; Miller and Stapleton, 1989; Szabo and Fegyverneki, 1995). The results are shown in Figures 3 and 4. The mean daily potential doses of PCP, 2,4-D, and BaA through all routes compared to the excreted compounds/metabolites in urine are shown in Figure 3 in units of nmol/kg, so that no molecular weight correction factor need be applied. The potential dose and the amount excreted are similar for PCP and BaA, and are also reasonable for 2,4-D.

However, for CPF, shown in Figure 4, the excreted amount of its metabolite TCP is approximately five times the potential dose of CPF. If the amounts of TCP that are in the environmental media and especially in the food that the children consumed are taken into account, the explanation is clear. The TCP potential dose from dietary ingestion alone (liquid and solid food, mean intake dose 2.5 nmol/kg day) is more than sufficient to produce the amount of TCP in the urine (1.1 nmol/kg day excreted). TCP has been found often in other environmental media (Chuang et al., 1999a; Morgan et al., 2002; Wilson et al., 2002).

Urinary measurements of TCP have been shown to correlate well with CPF exposures for persons in agricultural settings where CPF has been applied recently (Fenske and Elkner, 1990). A recent longitudinal study of the urinary OP metabolite levels of children living in an area where OP pesticides are used in agriculture found that the metabolite concentrations were higher in months when OPs were used on orchards, despite no direct contact of family members with the pesticides or location near the sprayed orchards (Koch et al., 2002). In a study of CPF levels in house dust compared to urinary TCP levels for children of pesticide applicators, farm workers, and nonagricultural workers, urinary TCP did not match well the parental occupation, although it did match home garden pesticide use (Fenske et al., 2000, 2002b). Observed levels of TCP may not match well the potential intake of CPF in nonacute exposure settings. In a study of the urinary TCP levels for human volunteers in the NHEXAS-Maryland study *versus* dietary

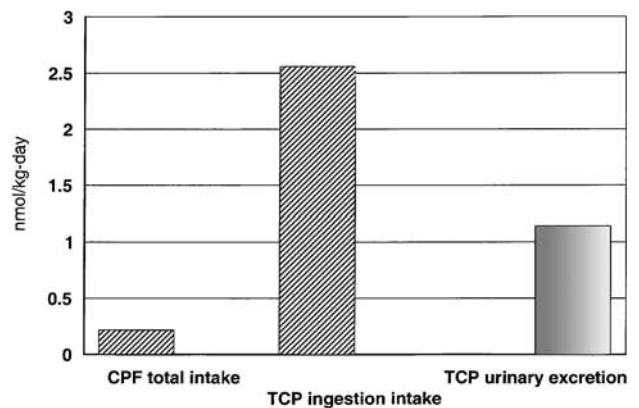


Figure 4. Mean aggregate daily PD (intake) of chlorpyrifos (CPF) through inhalation and ingestion, intake of 3,5,6-trichloro-2-pyridinol (TCP) through ingestion, and urinary excretion of TCP for nine children, in nmol/kg day.

intakes of CPF calculated from US FDA Market-Basket Surveys 1993–1997, the calculated dietary CPF accounted for only about 7% of the TCP (MacIntosh et al., 2001). At the low levels of exposure encountered by the children in their everyday environments in the study reported here, the measured levels of TCP in urine imply a false, high estimate of CPF exposure, as shown in Figure 4. Based on the findings in this study, it is suggested that the multimedia exposures of young children without recent acute exposures can be estimated from urinary measurements for PCP, 2,4-D, and BaA. However, it may not be possible to estimate their exposures to CPF by measuring TCP in their urine, because of high levels of TCP in food and other environmental media.

Conclusions

In this study of nine preschool children's aggregate exposures to a suite of persistent pesticides and other POP in their everyday environments, both at home and at their child day-care centers, there were five major findings. (1) The children's exposures were very low for most pollutants, with a few exceptions when pesticides had been applied recently in their environments. (2) Although there were slight and statistically significant differences in levels of some compounds between day care and home, and between low- and middle-income households, these differences and overall levels were small. Results for a larger number of participants are needed to draw meaningful, physically significant conclusions about these differences. (3) Except for the most volatile compounds, dietary ingestion was the primary route of exposure. Inhalation was the most important route for the organophosphate pesticides, the organochlorine pesticides, and the polychlorinated biphenyls, but for these groups of compounds, dietary ingestion was nearly as important.

(4) Urinary biomarker measurements agreed adequately with measured aggregate exposures for PCP, 2,4-D, and BaA and could be used as biomarkers of exposure to these compounds.

(5) Urinary biomarker measurements of the CPF metabolite TCP did not agree with the measured aggregate exposures to the parent CPF. Dietary ingestion of the pyridinol itself, even without contributions from other environmental media, produced urinary levels far higher than those that could have come from exposures to CPF. Therefore, in children without acute exposures to CPF, TCP is not a reliable biomarker of exposure.

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