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Perspiration increased human pesticide absorption following surface contact during an indoor scripted activity program

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Homeowners and professional applicators frequently use chemicals to control insect pests in urban environments. The identification and evaluation of determinants of human exposure are critical to conduct reliable and responsible human exposure assessments following indoor residential chemical applications. The effect of sweat on absorbed dose in humans was evaluated with human volunteers who participated in a structured activity program (SAP). Participants ($n = 20$) performed a warm-up exercise to induce light sweating prior to an SAP on chlorpyrifos(cp)-treated nylon carpet. Absorbed daily dosages (ADDs) were calculated using urinary biomonitoring of trichloropyridinol. In two separate exposures, participation in the warm-up exercise prior to the exposure SAP resulted in an increased ADD of CP equivalents by approximately 50%. Measured ADDs averaged 2.8 (SAP 1) and 2.0 (SAP 2) μg CP equivalents/kg/day in volunteers who participated in the warm-up exercise. In participants who rested prior to the exposures, ADDs were significantly lower at 1.9 (SAP 1) and 1.3 (SAP 2) μg CP equivalents/kg/day. Perspiration may also be a determinant of exposure in active children and field workers. Measured ADDs were less than estimates of ADD made from environmental measurements including CP deposition, the California roller, and clothing dosimeters worn by participants.
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Introduction

To clarify the extent of human exposure and the absorbed dose following pesticide applications, it is important to identify and evaluate determinants of exposure and bioavailability. Default assumptions used to calculate human exposure and absorbed daily dosage (ADD) from environmental measurements of air and surface residues typically overestimate measurements obtained through biomonitoring (Krieger et al., 2000). Published exposure estimates following the indoor use of foggers have been as high as 50 mg/kg/day (Berteau et al., 1989), while measurements of ADD following indoor pesticide use normally range from 1 to 10 µg/kg/day (Krieger et al., 2001). The difference between predicted and measured ADD warrants the study of potential determinants of exposure and bioavailability.

Moisture has been shown to increase chemical transferability from nylon carpet (Williams et al., 2002). Recent *in vitro* measurements of chlorpyrifos (CP) absorption following dosing with CP-treated nylon carpet fibers resulted in significantly higher chemical transfer to the skin surface when synthetic sweat was applied prior to dosing (Williams et al., manuscript). Skin moisture, as influenced by relative humidity, has also been shown to increase the dermal absorption of propoxur (Meuling et al., 1997). The correlations of both (1) transferable chemical residue with moisture (Williams et al., 2002) and (2) skin moisture with dermal absorption (Meuling et al., 1997) have been demonstrated independently, but the relationship between perspiration, potential human exposure, and ADD has not been studied in a setting that represents normal human contact with pesticide-treated indoor surfaces.

The organophosphate insecticide CP was chosen as a surrogate for semivolatile chemicals that are used indoors. CP products are no longer registered for indoor use as a result of an agreement between the major manufacturers and the USEPA (Browner, 2000). CP is a well-characterized compound with low mammalian toxicity that has been used extensively in human exposure studies (see Krieger et al., 2001). The CP biomarker, 3,5,6-trichloro-2-pyridinol (TCP) is stable, not appreciably stored in tissues, excreted in urine ($t_{1/2} = 27$ h, Nolan et al., 1984; $t_{1/2} = 30$ h, Griffin et al.,

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1999), and measurable at levels typically encountered and well below regulatory LOAELs or NOAELs.

A structured activity program (SAP) intended to represent indoor contact with treated surfaces following pesticide applications corresponding to more typical activities during the course of an entire day has previously been used to estimate human exposure potential (Ross et al., 1990, 1991; Krieger et al., 2000; Bernard et al., 2001). The SAP is controlled, reproducible, and can be used to evaluate and compare determinants of human exposure and absorbed dose. The ADD resulting from participation in the SAP is comparable with levels typically observed following normal chemical use (Krieger et al., 2000, 2001). For these reasons, the SAP was chosen for use in this study.

This study was designed to evaluate the effect of perspiration resulting from a moderate 10-min warm-up exercise on the ADD of CP during an SAP.

Materials and methods

Study Design

Healthy adult males were recruited by word of mouth from the university community for the SAP consistent with a protocol approved by the University of California, Riverside, Institutional Review Board (HS-01-004). Participants were briefed about study objectives and 21 participants provided informed consent. One participant was unavailable for the second SAP due to a personal commitment. The average weight and age of the participants was 76 ± 15 kg and 28 ± 3 years, respectively.

Participants were divided by random draw into two groups. Group 1 participated in a warm-up exercise. Group 2 was seated nearby during the warm-up. The warm-up exercise program was a 10-min set of cardiovascular Jazzercise® activities on an untreated carpet. Following the warm-up, all participants were randomly assigned SAP plots on treated carpets. Each participant completed a carefully controlled, 20-min SAP (Jazzercise®; Ross et al., 1990). After 2 weeks, in accordance with a crossover design, the SAP was repeated with group 2 participating in the warm-up exercise, while group 1 rested prior to the exposure period.

Study Site

These studies were conducted in Riverside, CA, in a 650 m^2 commercial building that had no recent history of pesticide use. The study area (10 m × 10 m) was carpeted with four HUD certified plush nylon carpets (weight ≥ 24 oz/yd^2, density ≥ 2600 oz/yd^3, and filament ≥ 12 denier; HUD, 1993). New carpets were used for each study. Each treated carpet was divided into two rows of three plots (1.3 m × 1.7 m) to be used by participants during the SAP and a central, undisturbed corridor (4m × 0.7 m) that was used for environmental sampling.

Chemical Application to Carpet

CP was applied to nylon carpets as an aqueous suspension of Dursban™ Pro (Dow AgroSciences LLC, Indianapolis, IN, USA; EPA Reg. No. 62719-166). A 0.5% suspension of Dursban™ Pro was applied for each study using a TeeJet® nozzle system (9503EVS; Spraying Systems Co.®, Wheaton, IL, USA) attached to a mobile cart (Bernard et al., 2001) that was pulled across the carpet at 0.55 m/s. The spray system was primed to remove air from spray lines and pressurized to 40 psi with nitrogen.

Environmental Sampling

Foil-backed cotton cloth coupons were placed in the aisles between the treated carpets to measure CP deposition ($\mu g/cm^2$). The sites were undisturbed following the CP application.

Transferable CP residue was measured following extraction of a cotton cloth rolled with a 30-pound short-handled modification of the CDFA roller (Ross et al., 1990; Bernard et al., 2001). The cloth, a prewashed 100% cotton sheet (35 cm × 35 cm; Gerber Childrenswear, Charlotte, NC, USA), was placed on the treated carpet and covered with roofing paper (Ratan Red Rosin Sheeting, Salinas Valley Wax Paper Co., Salinas, CA, USA) and a screen template. The roller was passed back and forth 10 times over the sampling unit.

Transferable chemical residue samples were collected prior to CP application and immediately following the SAP. Following the structured activity program, eight transferable chemical residue samples were collected from unused locations within the sampling corridors. An additional eight transferable chemical residue samples were collected from randomly chosen plots used by study participants during the SAP. Samples were transported in a cooler on reusable ice to the laboratory where they were stored in a freezer (−15°C) until analysis. Previous validation showed 104 ± 18% and 96 ± 3% recovery of CP from cotton cloth low (48 μg, $n = 5$) and high (482 μg, $n = 5$) spikes, respectively (Bernard, 2001).

The deposition coupons (250 ml) and cotton sheets (150 ml) were extracted with ethyl acetate for 20 min using an Eberbach shaker (1.5 in throw at 270 cycles/min). A portion of the extract was retained for CP analysis.

The transferable percent of total CP residue was calculated as follows:

$$\text{Transferable percent} = (\text{Transferable residue}/\text{CP deposition}) \times 100$$

Structured Activity Program

The warm-up exercise program was a 10-min set of cardiovascular Jazzercise® activity in an untreated area (“Cut you loose”, R1-99-17, 3 min 45 s; “Fire wire”, R1-02-6, 2 min 50 s; and “This is acid”, R1-02-8, 3 min 35 s; Jazzercise®, Carlsbad, CA, USA). The exposure SAP is a set

of 20-min, high surface contact, low impact Jazzercise® exercises and stretches (Ross et al., 1990; Krieger et al., 2000).

Participant Clothing Dosimeters

Each participant wore one 100% cotton sock (500 cm^2) on their right foot and a pair of 100% cotton shorts (3925 cm^2). Clothing was washed in a home washer with light laundry detergent followed by two additional wash cycles with warm water only. The clothing was dried using a home clothes dryer. No additional clothing was worn. Each person entered and exited the study site wearing surgical booties (Ulti-Med®,St Paul, MN, USA) to avoid contact with treated carpet. Following the SAP, the sock and shorts were collected and later extracted with ethyl acetate as indicated above. A portion of the extract was retained for analysis.

Using previous measurements of chemical transferred to cotton socks (26% of the total chemical transferred was retained by socks; Ross et al., 1990), dermal dose can be estimated. One sock represents one-half a sock pair or 13% of the total amount of CP transferred to a whole body dosimeter (Ross et al., 1990). Total chemical transferred is estimated as follows:

$$\text{Total chemical transferred } (\mu g) = \text{Amount on sock}(\mu g)/0.13$$

Sweat Collection

Each study participant wore a Macroduct® sweat collector (Wescor, Inc., Logan, UT, USA), on the volar surface of the left forearm approximately 15 cm from the wrist. The collector has a shallow concave undersurface with a small aperture that leads to microbore plastic tubing. Sweat flows by capillary action through the aperture and is retained in the microbore tubing. A small amount of water-soluble dye on the collector allows for visual gauging of collected sweat. Immediately following the SAP, the sweat level was marked on the microbore tubing. Volume determination was conducted by flushing sweat from the tubing and volumetrically refilling with dyed water to the mark using a blunt tipped syringe (100 μl; Hamilton Co., Reno, NV, USA).

Urine Collection

Each participant collected a morning urine specimen prior to the SAP and for 5 days following the SAP (Bernard, 2001). Samples were collected in polyethylene bottles, placed in home freezers, and brought to the laboratory on the final collection day. There were 21 and 20 participants who completed the SAP and subsequently provided urine samples for exposure periods 1 and 2, respectively. No other sources of CP exposures were reported.

CP and Biomarker Analysis

CP was measured from deposition coupons, transferable residue dosimeters, and participant clothing extracts by GLC-FPD using a CP standard curve with methylchlorpyrifos as an internal standard (Chemservice, West Chester, PA, USA) integrated by peak area. Samples were injected into a Hewlett Packard 5890 gas chromatograph equipped with an autosampler (Hewlett Packard 6890 series; Hewlett Packard, Wilmington, DE, USA) and an HP-5 fused silica column (30 m × 0.32 mm × 0.25 μm film; Hewlett Packard, Wilmington, DE, USA). Helium was used as a carrier gas at a flow rate of 2 ml/min. The injector was set to 275°C. The oven temperature started at 50°C for 1 min and increased at 20°C/min to 300°C, holding at 300°C for 3 min.

The urinary CP biomarker, TCP and creatinine concentrations were measured by Pacific Toxicology Laboratories (Chatsworth, CA, USA). Conjugated and unconjugated TCP was analyzed from acid hydrolysates of urine (Aston, 1998). Creatinine concentration was determined by the method of Jaffe et al. (1886) using a Beckman CX7 analyzer and Beckman Coulter Synchron CX® Systems Creatinine Reagent Kit (P/N 443340, Beckman Coulter, Inc., Fullerton, CA, USA).

CP equivalents excreted in urine were calculated based upon daily urinary creatinine as follows:

$$\text{CP equivalents} = \frac{\text{CP}_{\text{FW}}}{\text{TCP}_{\text{FW}}} \times \text{TCP}_{\text{excreted}}$$

$$\text{TCP}_{\text{excreted}} = \frac{\text{TCP}_{\text{urine}} \times \text{Daily creatinine excretion}}{\text{Creatinine}_{\text{urine}}}$$

The calculation includes a stoichiometric factor (ratio of formula weights of CP to TCP) multiplied by the creatinine-adjusted TCP concentration in urine. Daily creatinine excretion of 1.7 g/day was used (ICRP, 1975). The daily CP equivalents were summed and the cumulative CP equivalents excreted for 5 days following the SAP for each participant was used to determine ADD (μg/kg). No adjustments were made for background TCP in urine. TCP levels were 0.06 ± 0.05 μg/kg body weight and 0.07 ± 0.04 μg/kg body weight prior to exposures 1 and 2, respectively, giving no evidence of CP or TCP retention during the study period (see Figures 1 and 2).

The TCP excretion half-life was calculated graphically from the best-fit line of the semilogarithmic plot of TCP remaining to be excreted vs. time (Rowland and Tozer, 1980) (see Figures 3 and 4).

Transfer Coefficients

Measurements of ADD were used to calculate transfer coefficients (TC) using the USEPA algorithm for calculating human exposure following indoor residential applications on carpet. TCs were calculated from participants who did not complete the warm-up exercise as follows:

$$\text{TC} = \text{PDR}/(\text{ISR} \times \text{ET})$$

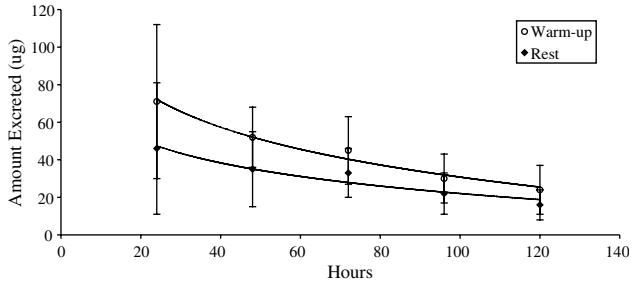

Figure 1. Excretion of CP equivalents following exposure 1. CP equivalents excreted (mean ± SD; see Table 2) following participation in an SAP 24 h after a broadcast spray application. The equation of the best-fit line is $y = -29.012\ln(x) + 164.38$ ($r^2 = 0.98$) and $y = -17.813\ln(x) + 104.06$ ($r^2 = 0.93$) for participants completing the warm-up exercise and resting, respectively.

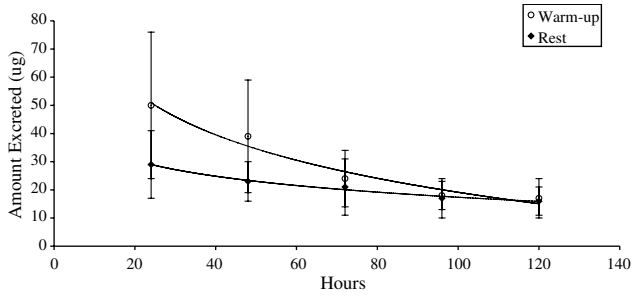

Figure 2. Excretion of CP equivalents following exposure 2. CP equivalents excreted (mean ± SD; see Table 3) following participation in an SAP 24 h after a broadcast spray application. The equation of the best-fit line is $y = -22.282\ln(x) + 121.75$ ($r^2 = 0.97$) and $y = -8.148\ln(x) + 104.06$ ($r^2 = 0.99$) for participants completing the warm-up exercise and resting, respectively.

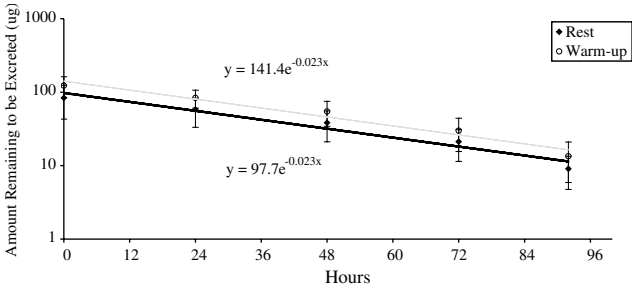

Figure 3. Semilogarithmic plot of amount of TCP remaining to be excreted *vs* time following exposure 1. Amount of TCP (mean ± SD) remaining to be excreted following participation in an SAP 24 h after a broadcast spray application. The $t_{1/2}$ calculated from the best-fit line was 30 h for both participants who completed the warm-up exercise and those who rested prior to exposure.

where TC is the transfer coefficient, PDR is the potential dose rate, ISR is the indoor surface residue (analogous to transferable surface residue), and ET is the exposure time. The PDR is calculated from measured absorbed dose (PDR = ADD/dermal absorption factor (9.6%)). Assuming that the ISR is 5% of the deposition rate (USEPA, 2001), the

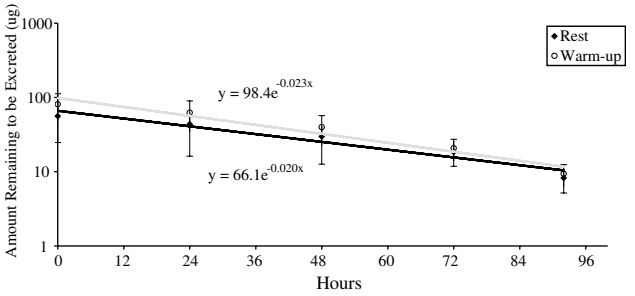

Figure 4. Semilogarithmic plot of amount of TCP remaining to be excreted *vs*. time following exposure 2. Amount of TCP (mean ± SD) remaining to be excreted following participation in an SAP 24 h after a broadcast spray application. The $t_{1/2}$ calculated from the best-fit line was 30 and 35 h for participants who completed the warm-up exercise and those who rested prior to exposure, respectively.

TC is 2700 and 1800 cm^2/h for exposures periods 1 and 2, respectively.

Statistical Analysis

Comparisons were made for CP deposition and transferable CP residue following the applications for exposure periods 1 and 2. Differences ($P < 0.05$) were determined using Student's *t*-test. A two-way analysis of variance (ANOVA), followed by least squares means, was conducted to compare total TCP excretion following exposure periods 1 and 2 to determine differences ($P < 0.05$) between the exposure day and participation in the warm-up exercise. Furthermore, an analysis of covariance (ANCOVA), followed by least squares means was conducted with total TCP excretion and sweat as a covariate ($P < 0.05$). The sample ($n = 30$) was restricted by eliminating participants from the warm-up group that had no measurable sweat and participants with measurable sweat from the resting group.

Results

Environmental Conditions and CP Measurements

Environmental conditions were similar on both study days. The temperature was 23.5°C and 25.5°C with 50% and 48% relative humidity for exposures 1 and 2, respectively.

The results of the CP deposition are reported in Table 1. The total CP residue measured from deposition coupons, 34.1 ± 3.9 and 31.4 ± 3.4 $\mu g/cm^2$ for exposures 1 and 2, respectively (mean ± SD), was similar on both days. Transferable surface residue was measured at undisturbed sites using the California roller following the SAP, approximately 24 h following the CP application. Undisturbed sites in the sampling corridor were 0.25 ± 0.07 and 0.23 ± 0.07 $\mu g/cm^2$ for exposures 1 and 2, respectively (Table 1). Used plots had 0.16 ± 0.10 and 0.16 ± 0.04 $\mu g/cm^2$ for exposures 1 and 2, respectively (Table 1).

Transferable CP residue, measured in the environmental sampling corridor using the modified California roller

Table 1. Environmental CP measurements following broadcast spray applications of 0.5% Dursban™ Pro on medium pile nylon carpet.

		Transferable residue ($\mu g/cm^2$)	
	Total CP[a] ($\mu g/cm^2$)	Unused[b]	Used plots[c]
Exposure 1	34.1 ± 3.93	0.245 ± 0.072^{A}	0.155 ± 0.096^{C}
Exposure 2	31.4 ± 3.38	0.230 ± 0.068^{AB}	0.160 ± 0.038^{BC}

[a]Total CP residue measured from foil-backed, cotton cloth deposition coupons.
[b]Transferable CP residue measured using the modified California roller 24-h following application from undisturbed plots.
[c]Transferable CP residue measured using the modified California roller 24-h following application from plots used for the SAP.
Significant differences ($P<0.05$) are indicated by letters *A*, *B*, and *C*; values with the same letter are not different (ANOVA, least-squares means; SAS).

method, was 0.7% of the CP deposition rate for both the exposure periods. This range for transferable residue from carpet is comparable with values of 1–3% reported by Krieger et al. (2001), Ross et al. (1991), and Bernard (2001) following indoor carpet treatments.

Clothing as an Exposure Dosimeter

Clothing dosimeters also retained CP. The mean CP removed by socks was 1281 ± 541 and 368 ± 168 μg for exposures 1 and 2, respectively. Using the sock dosimeter and the Ross et al. (1990) distribution data, the mean calculated amount of CP transferred during the SAP was 9900 ± 4200 and 2800 ± 1300 μg/person for exposures 1 and 2, respectively. The mean CP transferred to shorts was 2311 ± 1071 and 1144 ± 539 μg for exposures 1 and 2, respectively.

Sweat Collection

Participants who completed the warm-up exercise perspired lightly prior to exposure. Measurable sweat was collected in the Macroduct collectors from eight and 12 participants for exposures 1 and 2, respectively. Sweat collected from participants who rested prior to exposure was 2.7 ± 9.0 and 5.3 ± 9.8 μl following exposures 1 and 2, respectively. Participants who completed the warm-up prior to exposure yielded 35.4 ± 29.6 and 38.0 ± 27.5 μl following exposures 1 and 2, respectively. The amount of sweat collected (μl) from the forearm was a significant covariate with absorbed dose.

Biomarker Analysis

All urine samples were received from participants during the 6-day collection periods. Compliance was particularly high due to recruitment of participants from the graduate student population. TCP in background samples was less than 10% of the cumulative TCP excreted following the exposure periods. Prestudy TCP levels ranged from less than the limit of quantification (2 μg/l; $\approx$ 0.05 μg/kg) to 0.09 μg/kg, quantifiable samples averaged 0.04 μg/kg (>75%). Samples were not adjusted for background TCP excretion. Cumulative TCP excretion over the 5-day postexposure period (Figures 1 and 2) was 103 ± 44 and 69 ± 27 μg following exposures 1 and 2, respectively. The TCP excretion $t_{1/2}$ was about 30 hours in each exposure (Figures 3 and 4).

Participants who completed the warm-up exercise prior to the exposure period excreted significantly more TCP than participants who rested prior to the exposure period on both study days. TCP excretion was 123 ± 39 and 88 ± 32 μg (2.8 and 2.0 μg CP equivalents/kg/day) for persons who warmed-up prior to exposures 1 and 2, respectively. Average TCP excretion was 84 ± 41 and 57 ± 22 μg (1.9 and 1.3 μg CP equivalents/kg/day) for participants who rested prior to the exposure period following exposures 1 and 2, respectively. TCP excretion stoichiometrically converted to CP exposure equivalents are reported in Tables 2 and 3.

Table 2. Daily CP equivalents (μg) from urine biomonitoring for exposure 1.

	Control						
	Day 0	Day 1	Day 2	Day 3	Day 4	Day 5	$\sum$ Days 1–5
Warmed-up participants							
1	11	62	46	51	34	32	225
3	9	28	30	26	19	26	130
5	26	94	61	70	21	13	260
6	3[a]	71	49	38	23	11	191
7	2[a]	24	28	27	17	11	107
9	3[a]	72	62	58	28	21	241
12	19	157	75	46	28	23	329
14	5	108	60	60	39	37	303
15	15	34	40	57	61	53	245
17	7	60	68	15	26	16	185
Mean	10	71	52	45	30	24	222
SD	8	41	16	18	13	13	70
Rested participants							
2	12	81	28	28	22	21	180
4	18	103	75	46	38	16	277
8	6	40	34	32	20	15	142
10	2[a]	24	24	19	11	9	88
11	9[a]	104	55	43	33	21	256
13	3[a]	34	36	27	18	16	131
16	6	47	11	49	35	29	172
18	10	6	38	28	18	15	105
19	12	35	56	46	28	27	192
20	2[a]	16	20	11[b]	7	4	58
21	3	18	13	12	9	6	57
Mean	8	46	35	33	22	16	151
SD	5	35	20	13	11	8	73

[a]TCP concentration was less than the limit of quantification (LOQ = 2 μg TCP/l). One-half of the LOQ was used for the calculation.
[b]Second void used in absorbed dose calculation.
Total CP equivalents were calculated from TCP excreted in morning urine specimens. Samples were collected prior (control) and for 5 days following the exposure period with urine volume adjusted based upon urinary creatinine concentration (1.7 g/day; ICRP, 1975).

Table 3. Daily CP equivalents (μg) from urine biomonitoring for exposure 2.

	Control						
	Day 0	Day 1	Day 2	Day 3	Day 4	Day 5	$\sum$ Days 1–5
Warmed-up participants							
2	1[a]	89	38	33	23	12	195
4	2[a]	82	44	38	21	17	201
8	12	32	27	13	20	16	108
10	2[a]	16	11	10	8	8	53
11	2[a]	49	51	31	21	27	177
13	14	37	25	14	13	8	98
16	14	22	23	18	15	12	89
18	22	49	42	17	17	22	146
19	12	80	85	32	23	29	249
20	4	48	43	32	21	15	158
Mean	9	50	39	24	18	17	147
SD	7	26	20	10	5	7	60
Rested participants							
1	2[a]	32	33	16	27	22	130
3	5	25	23	25	23	13	109
5	11	25	18	10	13	8[a]	73
6	9	27	20	20	11	8	86
7	5	16	12	5	8	8	49
9	11	19	21	35	14	11[b]	100
12	14	59	30	23	12	17	141
14	6	31	27	35	15	21	129
15	9	37	29	24	27	18	137
17	7	23	16	16	18	18	90
Mean	8	29	23	21	17	16	104
SD	4	12	7	10	7	5	30

See Table 2 footnote.

Discussion

Contact with pesticide-treated carpet during the SAPs resulted in low levels of CP exposure. The aggregate TCP background observed here is consistent with previously reported values by Hill et al. (1995; $<$LOD to 2 μg/kg) and Krieger et al. (2001; 0.1–1 μg/kg). ADD ranged from 0.6 to 3.6 μg CP equivalents/kg in the 5-day period following the SAP exposure. CP can safely be used at levels that are typically encountered following indoor applications in experimental human exposure studies (see Table 4).

Perspiration and ADD

When participants completed the warm-up exercise prior to the exposure period, they absorbed significantly more CP based upon TCP excretion during the study period. The ADDs were 222 ± 70 vs. 151 ± 73 μg CP equivalents following exposure 1 and 147 ± 60 vs. 104 ± 30 μg CP equivalents following exposure 2 (Tables 2 and 3). The warm-up increased contact-transfer and absorption at CP levels similar to those that are typically encountered following indoor residential use. The 10-min warm-up period consisted of medium to heavy Jazzercise® cardiovascular conditioning exercises performed while standing. The warm-up period induced physiological responses consistent with light exercise, such as increased perspiration, heart rate, body temperature, circulation, and tissue perfusion, which may all have influenced absorption. Persons involved in vigorous activities may have a higher exposure potential, due to both physical (e.g. increased number of surface contacts) and biological considerations (e.g. dermal absorption), than persons who are inactive. This may be especially true in indoors for children, who excreted more TCP (μg/kg-day) than their parents during situational monitoring studies in California residences (Krieger et al., 2001).

Based upon the findings presented here, moisture is an important determinant of human exposure that results in increased absorbed dose. The increase of percutaneous absorption measured *in vitro* was not associated with an increase in the percent or rate of absorption (Williams et al., manuscript), suggesting that moisture increases the initial chemical transfer and skin loading providing a larger chemical reservoir for subsequent absorption. For conditions where moisture is present, from physiological (e.g., work), biological (e.g., sweat, saliva, etc.), or environmental (e.g., rain, heat, relative humidity, formulation, etc.) sources increased human exposure potential exists.

Environmental CP Measurements and ADD

Environmental measurements, including CP deposition and transferable CP residue, were also used to evaluate the relationship between environmental measurements and exposures. ADDs calculated from environmental measurements overestimated ADDs measured through urine biomonitoring.

Uniform applications in experimental studies are an important means to reduce the variability of ADD and to increase the reliability of measurements of chemical persistence, transferability, and potential human exposure. Surface deposition levels of CP (μg/cm^2) following discharge from fogger canisters are unevenly distributed, with levels typically observed as a declining gradient from the source, ranging from over 17 μg/cm^2 to less than 1 μg/cm^2 (Krieger et al., 2000, 2001). In the present study, applications were made using a mobile spray cart (Bernard et al., 2001) to deliver reproducible and uniform CP deposition. The CP deposition rates of 34 ± 3 and 31 ± 3 μg/cm^2 for the two exposure periods were not significantly different ($P > 0.05$) over the entire study area. This created a similar study environment on both days that is representative of levels that may be encountered indoors.

Transferable residues are advocated in indoor residential standard operating procedures (USEPA, 1997b) to estimate dose. Absorbed dose was estimated using the transferable CP residue measured with the California roller, 9.6% for dermal

Table 4. Calculation of CP daily dosage using four sampling methods.

Method	*Exposure 1 (μg/kg)*		*Exposure 2 (μg/kg)*	
	Calculated	Measured	Calculated	Measured
Cotton cloth and CDFA roller[a]	14	—	13	—
Personal short dosimeter[b]	15	—	7	—
Personal sock dosimeter				
Residue transfer is even over entire body[c]	64	—	17	—
Residue transfer to body regions based on WBD[d]	13	—	4	—
Urine biomonitoring[e]	—	1.9	—	1.3

[a]CP transferred to (cotton cloth [$\mu g/cm^2$] × body surface area [cm^2])/body weight (kg).
[b]CP transferred to (shorts [$\mu g/cm^2$] × body surface area [cm^2])/body weight (kg).
[c]CP transferred to (sock [$\mu g/cm^2$] × body surface area [cm^2])/body weight (kg).
[d]13% sock contribution to whole body dosimetry chemical transfer (Ross et al., 1990); CP transferred to sock (μg) × factor for total chemical transferred to participant/body weight (kg).
[e]CP equivalents measured as TCP from urine biomonitoring.

CP absorption/day (Thongsinthusak, 1991), and assuming that contact during the SAP represents day-long surface contact (Krieger et al., 2000). The estimated absorbed dose was 14 and 13 $\mu g/kg/day$ for exposure periods 1 and 2, respectively (e.g. $(0.245\,\mu g/cm^2 \times 43{,}000\,cm^2/day \div 76\,kg) \times 0.096 = 14\,\mu g/kg/day$). Measurements of transferable CP residue made with the California roller were more predictive of measured absorbed dose than nominal application rates or total chemical residue on carpet when used with existing USEPA algorithms and transfer coefficients (Krieger et al., 2000).

Although the environmental measurements including the deposition rate and transferable chemical residue measured with the California roller were similar on both days, differences in transferable chemical residue retained by clothing dosimeters and ADD occurred between the two study days. No clear causes of the discrepancy between the studies are apparent. A measure of conditioning or fitness was not recorded, but some subjects reported minor muscle soreness following the first exposure period, which may have led to less intense activity during the second exposure period. The differences in transferable chemical residue retained by clothing and ADD from exposures 1 and 2 exist nominally, but are of similar magnitude and represent the trend previously seen in the relationship between environmental measurements and ADD.

In many cases biodynamic data are not available to support biomonitoring. In the present study, personal dosimeters were worn to provide a passive measure of a participant's exposure to treated carpet. Transferable CP residue measured from passive dosimetry can also be used to estimate potential dermal exposure. Estimates of potential dermal exposure from shorts were based upon CP transfers of 0.6 and 0.3 $\mu g/cm^2$ to the clothing. Assuming uniform chemical transfer over the entire body surface (19,400 cm^2; USEPA, 1997a) and including 9.6% for dermal absorption (Thongsinthusak, 1991), the ADDs for participants during exposure periods 1 and 2 were 15 and 7 $\mu g/kg/day$, respectively.

The dermal dose calculated with the amount of CP extracted from socks (2.6 and 0.7 $\mu g/cm^2$) retained in exposures 1 and 2 was 64 and 17 $\mu g/kg/day$, respectively. Using the dermal absorption factor and the Ross et al. (1990) observation that 26% of the dermal dose was transferred to socks, the resulting ADD was 13 and 4 $\mu g/kg/day$ for exposures 1 and 2, respectively. The differences in ADDs estimated from personal dosimetry are most likely due to uneven chemical transfer and distribution with respect to body region. Regions such as hands and feet (sock dosimeter) have greater surface contact than other body parts. Estimates made from personal dosimetry that include regional distribution data (13 and 4 $\mu g/kg/day$) probably provide the best estimate of the biomonitoring results.

Estimates of potential human exposure made from environmental sampling frequently overestimate ADD (Krieger et al., 2000). Uncertainties about distribution, duration of exposure, and individual activity patterns may be minimized by biomonitoring. The ADD of CP equivalents measured from urinary TCP excretion by persons who did not participate in the warm-up exercise were 1.9 and 1.3 $\mu g/kg/day$ for exposures 1 and 2, respectively. All estimations of ADD from environmental sampling overestimated the results measured from biomonitoring (Table 4). Dosimeters are a larger reservoir than skin with respect to pesticide transfer (Ross et al., 1990; Krieger et al., 2000). Even so, extrapolations of ADD directly from measurements of chemical transferability improve estimations of ADD made from nominal application rates or measured deposition rates (Krieger et al., 2000). These studies are limited by the use of CP as a surrogate for other semivolatile chemicals used indoors that might not be suitable for biomonitoring. Further evaluation of the relationship between chemical transferability and measured ADD is important to clarify other

determinants of exposure since such small fractions of total residue are available for transfer.

The TCs calculated from these exposure periods (2700 and 1800 cm^2/h) are much lower than those suggested by the USEPA (43,000 cm^2/hr; USEPA, 1997b) or even the more recent recommended values (14,500 cm^2/h; USEPA, 2001). The difference could occur for several reasons including the duration of exposure and assumptions regarding transferable chemical residue. The Jazzercise® exposure model represents day-long indoor exposure (16-h; Krieger et al., 2000), but the exposure rates of the 20-min exercise period have been extrapolated to longer periods by USEPA. This practice is inconsistent with results of situational exposure monitoring that have reported similar experimental and monitoring results (Krieger et al., 2001). The transferable CP residue measured here was approximately 1% of the surface deposition, also much lower than the recommended default value of 5% (USEPA, 2001). Whether default values are justified for the many and varied indoor activities that may result in human exposure is not established. Further evaluation of the relationship between situational monitoring (Krieger et al., 2001) and data sets developed experimentally will help clarify the role of TCs and may ultimately improve the accuracy of algorithms to estimate ADDs.

Participation in a warm-up activity prior to exposure significantly increased the ADD of CP. Continued evaluation in residential and occupational settings of the relationship between increased activity, sweat, and ADD of semivolatile chemicals used for pest control is necessary to elucidate the mechanism and develop predictive methodology to use these findings appropriately to estimate human exposure to chemical use.

The magnitude and range of ADDs measured in this study are comparable to previous SAPs conducted with CP following indoor fogging (Krieger et al., 2000), following an indoor broadcast spray (Bernard, 2001), from situational exposure biomonitoring (Krieger et al., 2001), and from treated turf (Bernard et al., 2001). This similarity is likely due to low chemical transferability of CP to humans during contact, relative to amounts retained by treated residential surfaces. The Jazzercise® SAP provides a method to evaluate determinants of human exposure that is representative of levels typically encountered following normal pesticide use.

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