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## **Personal Exposure to Polycyclic Aromatic Hydrocarbons in Appalachian Mining Communities**

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## **Abstract**

Coal mining activities may increase residential exposure to polycyclic aromatic hydrocarbons (PAHs), but personal PAH exposures have not been studied in mining areas. We used silicone wristbands as passive personal samplers to estimate PAH exposures in coal mining communities in Central Appalachia in the United States. Adults ( $N = 101$ ) wore wristbands for one week; 51 resided in communities within approximately three miles of surface mining sites, and 50 resided 10 or more miles from mining sites. Passive indoor polyurethane foam (PUF) sampling was conducted in residents' homes, and a sample of 16 outdoor PUF samples were also collected. Nine PAH congeners were commonly detected in wristbands (mean  $\pm$  standard deviation), including phenanthrene (50.2  $\pm$  68.7 ng/g), benz[a]anthracene (20.2  $\pm$  58.2 ng/g), fluoranthene (19.4  $\pm$  24.1  $ng/g$ ) and pyrene (15.2  $\pm$  18.2 ng/g). Controlling for participant characteristics and season, participants living closer to mining sites had significantly higher levels of phenanthrene, fluorene, fluoranthene, pyrene and ∑PAHs in wristbands compared to participants living farther from mining. Indoor air showed no significant group differences except for pyrene, but outdoor air showed significant or marginally significant differences for phenanthrene, fluorene, pyrene and

PAHs. The results suggest that mining community residents face exposure to outdoor miningrelated pollutants, and demonstrate that personal silicone wristbands can be deployed as effective passive sampling devices.

## **GRAPHICAL ABSTRACT**

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Conflict of Interest

The authors declare no competing interests.



#### **Keywords**

polycyclic aromatic hydrocarbons; Appalachia; coal mining

## **INTRODUCTION**

Surface coal mining operations generate air and water pollution that may impact the health of the public in surrounding communities (Bernhardt et al. 2012; Ghose and Majee 2007; Huertas et al. 2012; Kurth et al. 2015; Palmer et al. 2010). Surface mining activities include the use of explosives containing fossil fuels and the regular use of heavy diesel machinery including draglines, trucks, trains, conveyors and other equipment. Large quantities of coal, rock and soil are exposed and transported in the mining process, raising levels of particulate matter across the coarse to ultrafine size range (Ghose 2007; Kurth et al. 2014). Epidemiological studies document adverse public health conditions in coal mining communities in ways consistent with exposure to mining-related pollution (Cortes-Ramirez et al. 2018; Fernández-Navarro et al. 2012; Liao et al. 2010).

Polycyclic aromatic hydrocarbons (PAHs) are a class of chemicals originating from incomplete combustion of fossil fuels, wood, garbage or tobacco (CDC 2019). They are also present in un-combusted coal and other fossil fuels. Human exposure to PAHs may occur through inhalation, ingestion or dermal absorption and is known to increase cancer risk (Boström et al. 2002; K-H Kim et al. 2013). PAH exposures are also related to cardiovascular disease, skin conditions, and kidney and liver damage (Ayman Alhamdow et al. 2017; K-H Kim et al. 2013), and adversely affect fetal growth and child neurodevelopment (Perera et al. 2005; Perera Frederica et al. 2012). There is some evidence that PAHs in ambient environments are elevated in mining communities relative to nonmining control sites from coal itself and from the incomplete combustion of fossil fuels used in mining operations as measured by outdoor air sampling (Kurth et al. 2015). The current

study extends prior research by examining whether personal exposures to PAHs are also present at higher levels among residents of mining communities compared to residents of non-mining communities.

Several methods have been developed to measure personal exposure to environmental contaminants (Asbach et al. 2017; Bohlin et al. 2007; Dixon et al. 2018; Jantunen et al. 2002; O'Connell et al. 2014). The current study employs personal silicone wristbands. Wristbands are small, unobtrusive and require no external power source. Previous research on silicone wristbands show that they are practical, valid, readily accepted by participants, and are able to detect PAHs and other chemical classes (Dixon et al. 2018; Hammel et al. 2016; O'Connell et al. 2014; Romanak et al. 2019). They have been used to identify elevated PAH levels among residents who live near natural gas fracking (Paulik et al. 2018) or oil refineries (Rohlman et al. 2019), and to assess occupational exposure among asphalt workers (O'Connell et al. 2014). The current study used silicone wristbands for the first time to examine personal exposure to PAHs among persons who reside in communities proximate to surface coal mining activities. We also examined the relationships among the PAH concentrations in wristbands and in indoor and outdoor air using passive polyurethane foam (PUF) samplers.

## **MATERIALS AND METHODS**

#### **Participant recruitment.**

A total of 101 adults living in rural communities in the Central Appalachian portion of southern West Virginia, eastern Kentucky, and western Virginia were recruited into the study and completed data collection. One resident per participating household was included. Only one participant dropped out after recruitment, for a completion rate of 99% (101/102). The geographic area selected for sampling was characterized by surface coal mining activities. Participants were recruited from communities within approximately three miles of surface coal mining sites, or communities at least 10 miles from mining sites (controls). Mining was confirmed through visual observation and community informants. Selected communities were canvassed door-to-door by trained research personnel during daylight hours, and participants were contacted in person at their homes and invited to take part. There were eight total communities from which participants were recruited.

#### **Sample collection.**

Passive air samplers equipped with polyurethane foam (PUF) discs were deployed indoors for 24 hours per day for 28 consecutive days in each participant's home. The samplers were place in a frequently used room away from appliances or heat sources. Most common rooms were living rooms/dens (n=53), kitchens (n=25), dining rooms (n=13) or other (n=10). In approximately 15% of cases ( $N = 16$ ), an outdoor PUF sampler was placed in participants' yards for 28 days (the rationale for the smaller sample being that outdoor PUFs would not be greatly different between nearby households in the same communities). Each community contained from one to four outdoor PUFs. One resident in each of these homes wore a silicone wristband (20 cm \* 1.2 cm, 0.2 cm thickness, [www.24hourwristbands.com,](https://www.24hourwristbands.com) Houston, TX) on the dominant wrist for the first 7 days of PAS-PUF deployment. Data

collection occurred during spring, summer and fall in 2017 and spring 2018; samples were collected in summer ( $N = 45$ ) and non-summer (spring,  $N = 54$ ; fall,  $N = 2$ ).

PUF discs were pre-cleaned by Soxhlet extraction with hexane/acetone  $(1:1, v/v)$  for 24 hours. PUFs were dried in the fume hood covered in foil, with a small opening for solvent evaporation, and transferred to the sampling locations wrapped in aluminum foil and sealed in Ziploc bags. Prior to deployment, wristbands were pre-cleaned via Soxhlet extraction with a mixture of ethyl acetate and hexane (1:1, v:v) for 24 hours, followed by ethyl acetate and methanol mixture (1:1, v:v) for 24 hours, then covered with foil and allowed to dry in a fume hood with a small opening for solvent evaporation, wrapped with foil after drying, sealed in Ziploc bags, and stored in a −20°C freezer.

After collection, wristbands were stored in sealed amber jars and stored at −20 °C until extraction. The PUFs were retrieved, individually covered with aluminum foil and sealed in Ziploc bags, shipped to the laboratory, and stored at −20 °C until extraction.

Descriptive information collected from participants included age in years, sex, race (white, black, Hispanic or other), educational attainment, marital status (married or not), current smoking (yes/no), occupation as a coal miner, use of coal for home heating, collection season (summer/non-summer), and hours spent outdoors.

#### **Sample analysis.**

The methods used in this study for PUF analysis were previously published and only a brief description is included here (Stubbings et al. 2018; Venier et al. 2016; Vykoukalová et al. 2017; Shaorui Wang et al. 2019). PUFs were added to accelerated solvent extraction (ASE) cells (Dionex ASE 350, Sunnyvale, CA, USA), spiked with surrogate standards  $(d_{10}$ phenantherene and d<sub>10</sub>-pyrene), extracted using 50:50 hexane/acetone at 100 °C and 1500 psi for three, ten minute static cycles, and concentrated to 1 mL using Rapidvap (Labconco, Kansas City, MO) after solvent exchange to hexane. The concentrated extracts were purified on a 2 cm - 3.5% water-deactivated silica column. The column was eluted with 12 mL of hexane, followed by 12 mL of a hexane/dichloromethane (DCM) mixture (1:1, v/v). All the fractions were blown down to 1 mL with  $N_2$  and spiked with known amounts of internal standards ( $d_{12}$ -perlyene,  $d_{12}$ -benz[a]anthracene, and  $d_{10}$ -anthracene). Both hexane fraction and hexane/DCM fraction were analyzed for PAHs.

Wristbands were spiked with surrogate standards mentioned above and ultrasonicated for two hours twice with 30 mL hexane/acetone (1:1, v:v). The extracts were combined, concentrated and blown down with  $N_2$ . The extracts were cleaned on a multi-layer column containing from bottom to top neutral alumina (3% deactivated), neutral silica (3% deactivated), Florisil (3% deactivated), and sodium sulfate, using 40 mL DCM as the eluent. The eluents were concentrated to 1 mL with  $N_2$  and spiked with internal standards mentioned above. Details on the specific protocols can be found elsewhere (Romanak et al. 2019).

PAHs were analyzed using an Agilent 6890 series GC coupled to an Agilent 5973 MS operated in the electron impact mode. A pulsed splitless injection of 1 μL at 285°C was used

to introduce the sample to the GC column and high purity helium (99.999%; Indiana Oxygen, Indianapolis, IN) was used as the carrier gas at 1.5 mL/min. The column used to attain GC resolution was a  $30 \text{ m} \times 0.25 \text{ mm} \times 0.25 \text{ mm}$  DB-5MS Ultra Inert capillary column (Agilent Technologies, Santa Clara, CA). The GC oven temperature was held at 70°C for 3 min, increased to 280°C at 30°C/min and held for 6 min, then to 300°C at 30°C/min and held for 12 min (total runtime 28.7 min). The GC/MS transfer line, ion source, and quadrupole temperatures were set at 285°C, 230°C, and 150°C, respectively (Romanak et al. 2019).

#### **Quality Assurance/Quality Control.**

Clean PUFs and wristbands were used as laboratory blanks. Field blanks, including those for PUFs and wristbands, were collected for each community and represented 15% of samples. Table S1 summarizes PAH levels in wristband and PUF blanks. The average surrogate recoveries were 83  $\pm$  19 % in PUFs and 106  $\pm$  26% in wristbands for d<sub>10</sub>-phenanthrene, and 84  $\pm$  17 % in PUFs and 101  $\pm$  27% in wristbands for d<sub>10</sub>-pyrene.

#### **Data Analysis.**

Descriptive summaries of study variables were calculated. Wristbands were usually collected after seven days but scheduling logistics on occasion resulted in a sampling range of six to nine days. Chemical levels in the wristbands were measured in  $\frac{ng}{g}/\frac{days}{w}$  worn)\*7 to adjust for the variability in sampling days which is within the linear uptake phase (Hammel et al. 2018; Kile et al. 2016). Similarly, indoor and outdoor PUFs were usually deployed for 28 days, and levels were measured as  $((pg/m<sup>3</sup>)/sampling$  days), the sampling rate for indoor and outdoor PUFs were 2.9 and 4 m<sup>3</sup>/day (Venier et al. 2016; Shaorui Wang et al. 2019).

Mean and median levels of chemicals detected from wristbands, indoor air and outdoor air were calculated overall, and separately for the mining and non-mining groups. Group differences on demographics and PAH levels were tested using chi-square tests for categorical variables or two-tailed t-tests for continuous variables unless otherwise noted. We examined bivariate Pearson correlations between wristband results and results from outdoor air, linking each participant to the outdoor PUF deployed within their community closest in time to their use of the wristband. Differences in total PAH levels by group (mining vs control), season (summer vs non-summer) and their interactions were examined using F tests. Multiple linear regression models were estimated using the chemical level as the dependent variable, and mining or control group as the independent variable, with covariates including season (summer or non-summer), participant age, sex, smoking, education, and use of coal in the home. Associations were considered significant at  $p < 0.05$ and marginally significant at  $0.05 < p < 0.1$ . Statistical analyses were performed using SAS software version 9.4 (Cary, NC).

## **RESULTS AND DISCUSSION**

#### **Participant characteristics.**

Table 1 presents a summary of participant characteristics. None of the participants were working as coal miners. The mining and control groups were not significantly different on sex, race (simplified to white versus other due to small numbers), marital status, current smoking, and use of coal as a home heating source. The mining group on average had less education, and was older. The mining group also wore the wristbands on average for a shorter time because of the sampling logistics. There were relatively fewer samples taken during the summer (versus spring or fall) in the mining group. Because of group differences, education, age, and season were selected as covariates for subsequent regression models. Although they were not different between groups, participant sex, smoking, and use of coal in the home were also included as potentially important covariates.

#### **PAHs in wristbands.**

Mean total PAH concentrations ( $PAHs$ , the sum of all PAH congener concentrations) were not significantly different based on sex, marital status, smoking, education, or age (results not shown). Table 2 provides a summary of PAH levels detected in wristbands for all participants. PAHs were detected in every wristband. Phenanthrene was the most frequently detected and abundant PAH (mean=50.2 ng/g, SD=68.7). Other abundant PAHs included fluoranthene (mean=19.4 ng/g, SD=24.1), fluorene (mean=8.7 ng/g, SD=7.4), pyrene (mean=15.2 ng/g, SD=18.2), and benz[a]anthracene (mean=20.2 ng/g, SD=58.2).

Fluoranthene and pyrene concentrations were significantly higher in the wristbands worn by mining group participants compared to the control. Phenanthrene, fluorene and benz[a]anthracene were also present on average at higher levels in the mining group than in the control, but differences were not significant.

Mean  $PAH$  levels were non-significantly higher in summer vs. non-summer ( $p < 0.29$ ) (Table S2). However, this table also shows that the interaction of mining and summer was significant: the highest PAH levels were detected in summer in the communities closer to mining. These results were corrected for the number of days wristbands were worn but not for covariates. We included summer season as a covariate in subsequent regression analyses.

Controlling for covariates, residence closer to mining was associated with significantly higher PAH levels for fluoranthene, fluorene, phenanthrene, and pyrene (Table 3). The total PAH level was also significantly higher in the mining environment. We ran a sensitivity analysis and retained as covariates only age, education and season and the results (not shown) were essentially unchanged.

Silicone wristbands capture both air and dermal exposures that mainly come from the back of the hand or palms (S. Wang et al. 2019) while PUFs only capture air exposure. The restriction of the PAH differences largely to wristbands and outdoor samplers, but not indoor samplers except for pyrene, might be the result of higher outdoor levels of these chemicals in mining-impacted communities. That is, the higher PAH levels detected among mining community residents may reflect greater inhalation and dermal deposition exposures from

outdoor sources. However, correlations between wristbands and outdoor air were generally low; wristbands might reflect exposures more accurately to the extent that they represent the time and space occupied by participants, whereas outdoor air in the current study were selected to represent the closest match in space and time to the participant, but may be temporally and spatially removed from participants' experiences. This may indicate that PAH levels in these environments fluctuate over time, perhaps in relationship to variation in short-term mining activity.

There are a limited number of previous studies that have investigated PAH exposures using silicone wristbands. Paulik et al. (Paulik et al. 2018) reported that PAHs from wristbands were significantly higher among persons who lived closer to, or worked at, natural gas extraction well sites. Rohlman et al. (Rohlman et al. 2019) used wristbands to study PAH levels among Native American persons living close to oil refineries. O'Connell et al. (O'Connell et al. 2014) used silicone wristbands and detected high levels of PAHs among an occupationally exposed group of asphalt workers. Other studies have used silicone wristbands to detect PAHs along with other chemical classes in a sample of urban and rural adults in Peru (Bergmann et al. 2017) or to detect PAHs among urban-dwelling pregnant women (Dixon et al. 2018).

Detection frequencies for PAHs common to our study occurred at higher rates relative to the study in Peru even though wristbands in our study were worn for one week versus one month in Peru (Bergmann et al. 2017), but were less frequently detected in our rural sample relative to the sample of urban women reported by Dixon et al. who wore wristbands for only two days (Dixon et al. 2018). The Paulik et al. (Paulik et al. 2018) study reported relatively higher PAH detection frequencies and quantities for their wristband study of natural gas extraction well sites; however, their small sample  $(N = 19)$  included not only people with well sites on their properties but also people who worked at the sites, whereas our study included people residing within several miles of mining sites, and included no one with occupational exposure. Levels of PAHs detected in wristbands in our study were generally higher than those reported for those living close to oil refineries (Rohlman et al. 2019).

#### **PAHs in indoor air.**

There was no significant difference between the two groups (mining and control) in types of rooms where the samplers were placed (chi-square=12.19, df=3, ns). Controlling only for length of deployment, PAHs detected from indoor air were not significantly different in the mining and control samples, with the exception that benz[a]anthracene was significantly higher in the mining group ( $p < 0.04$ ) (Table S3). No other differences were significant and the variability across samples was high; point estimates for total PAHs and for three-ring PAH congeners were non-significantly higher in the control group, whereas chrysene and pyrene were non-significantly higher in the mining group. Controlling for covariates in multiple regression models, differences between groups remained non-significant with one exception: levels of pyrene in the indoor air were significantly higher in the mining group (coefficient = 437.4, standard error = 196.2,  $p < 0.03$ ).

#### **PAHs in outdoor air.**

The results from the 16 outdoor air samples indicated that amounts of the same PAHs that were higher in the wristbands in the mining group were also higher in the outdoor air (Table 4). With this small sample size, only pyrene and  $PAHs$  were significant at  $p < 0.05$ , and fluorene and phenanthrene were significant at  $p < 0.10$ . PAH concentrations were almost three times higher in the mining area. These figures were corrected for season and length of deployment. Table S3 shows that the highest concentrations for most PHAs were observed in the mining group in the summer, although the group x season interaction was not significant.

Pearson correlations between PAH levels in wristbands and outdoor air are summarized in Table 5. Correlations were relatively stronger for the PAHs detected at higher frequency and amount, but were generally low.

#### **PAH Pattern.**

Figure 1 shows the percent contributions of individual PAHs to the PAH concentrations measured in indoor and outdoor air, and wristbands. Phenanthrene was the dominant PAH across all three sampled media, with percent contributions to ∑PAH levels of 61%, 54% and 37% in indoor air, outdoor air and wristbands, respectively. The percentage of three-ring PAHs, including fluorene, phenanthrene, and anthracene, in indoor air and outdoor air were higher than that of four- or five-ring PAHs (85% vs 15% for indoor and 74% vs 26% for outdoor). However, wristbands had a more diverse contribution from several PAHs, with greater relative amounts of four- and five-ring PAHs including pyrene, chrysene, benz[a]anthracene, benzo[b]fluoranthene, and benzo[k]fluoranthene. The higher abundance of four- and five- ring PAHs in wristbands compared to air samples is probably based on different sampling abilities of wristbands and passive air samplers. Wristbands have been shown to be effective for sampling both gas-phase and particle-phase PAHs as well as PAHs deposited on the skin, while PUFs are mostly effective for capturing gas-phase PAHs (S. Wang et al. 2019). With the exception of pyrene, which the EPA has not classified as to carcinogenicity, these other four chemicals are recognized by the EPA as probable human carcinogens (ATSDR 2013). Chronic exposure to PAHs, including to PAH mixtures, is also recognized as a threat to pulmonary, gastrointestinal, renal, reproductive, and dermatologic health (A. Alhamdow et al. 2017; ATSDR 2013; KH Kim et al. 2013).

#### **Discussion and limitations**

Results of the study indicate that persons residing in rural communities near surface coal mining operations in Appalachia, compared to people in other nearby rural communities farther from mining, had higher levels of personal exposure to PAHs as measured in silicone wristbands. PAH concentrations were significantly higher, as were levels of fluoranthene, fluorene, phenanthrene, and pyrene among the residents of mining communities. Similarly, based on 16 outdoor passive PUF samplers, outdoor PAHs were higher or marginally higher in communities closer to mining for PAHs and for fluorene, phenanthrene, and pyrene. Differences in PAH levels between mining and non-mining groups were not found for indoor air with the exception of pyrene.

Higher PAH levels in mining environments included both three- and four-ring compounds. Three-ring compounds are more likely to occur in vapor phase and may partially represent non-combustion sources, such as coal itself, whereas four-ring compounds are more likely to partition between vapor and particulate phases and result from incomplete combustion of fossil fuels (Choi et al. 2010; Krugly et al. 2014; K. Srogi 2007). Four-ring compounds have also been more clearly linked to health consequences and carcinogenic potential (Boström et al. 2002). In particular, the levels of four-ring PAHs fluoranthene and pyrene were almost double in mining communities than in control communities. We have previously demonstrated that wristbands integrate both inhalation and dermal exposures, and that the dermal exposure pathway represents a larger portion of the overall exposure captured by wristbands than inhalation (S. Wang et al. 2019). Human health studies on inhalation or dermal exposure to fluoranthene have not been reported but animal studies indicate that exposure results in liver and kidney damage (EPA 2012), and its chemical similarity to other PAHs suggests that it may have carcinogenic potential. Toxic effects of pyrene inhalation to a number of organ systems exists (NLM 2017).

Higher PAH levels were observed in summer in the mining group, for wristbands and for outdoor samplers. This may reflect seasonal variation in mining activity, or differences in meteorological conditions (e.g. inversions) that increase summertime exposures. The ∠PAH levels in mining environments from both wristbands and outdoor air were determined primarily from pyrene, phenanthrene, fluorene, flouranthene, and benz[a]anthracene, that is, from sources that may represent both non-combustion (i.e., coal) and combustion (i.e., diesel or other fossil fuels used in mining) origins and include compounds of health concern (CDC 2019; Choi et al. 2010). Uncombusted coal particles may introduce significant quantities of PAHs into the environment in areas characterized by coal mining, transportation and processing (Laumann et al. 2011). Masto et al. (Masto et al. 2019) found that phenanthrene, pyrene and flourene were the three most common PAHs detected in road dust in a coal mining area in India, and concluded that both coal dust and fossil fuel combustion were contributing sources. A study in a coal mining area in China (Yang et al. 2008) reported that pyrene, flouranthene and phenanthrene were among the highest PAH concentrations observed in soil.

Limitations of the study included the small sample of outdoor PUFs. Samples were collected mostly in spring, with relatively fewer summer samples, only two collected in fall season and no winter sampling. Length of time when wristbands were worn or PUFs deployed were not exactly equal between groups, but corrections were made for these differences. Actual mining activity can vary and it was not possible to determine how active a particular mine site may have been during data collection. The control group was at least 10 miles from active mining, but air pollution effects may extend beyond this range (Hudda et al. 2014; K. Srogi 2007; WHO 2006), which would render our group differences conservative. Whether differences in personal PAH exposures contribute to previously documented health disparities in mining communities is unknown. Correlations between wristbands and outdoor air were mismatched to the extent that deployment of PUFs did not match wristband deployment exactly in time, which may account for relatively lower correlations compared to those observed in other research (Paulik et al. 2018).

## **CONCLUSIONS**

The study demonstrates that previously documented (Kurth et al. 2015) elevations in outdoor ambient PAH levels found in coal mining communities were also present for personal exposures. Contact with PAHs may include both dermal and inhalation exposure routes. Exposures were highest in mining areas during the summer and likely include outdoor exposures to both coal itself and to incomplete fossil fuel combustion from mining activities. The study also extends the growing evidence base that silicone wristbands are of potential utility for personal exposure assessment of environmental chemicals of concern.

## **Supplementary Material**

Refer to Web version on PubMed Central for supplementary material.

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## **Highlights**

- **•** Living close to surface coal mining sites was associated with elevated PAH exposures
- **•** Differences were observed for silicone wristbands and outdoor PUFs, but not indoor PUFs
- **•** Personal silicone wristbands are practical and effective for passive sampling





Percent compositions of individual PAHs to total PAH contributions in indoor air, outdoor air, and wristbands.

#### **Table 1.**

Descriptive summary of study participants.



\* P values based on chi-square tests for categorical variables and two-tailed t-tests for continuous variables.

\*\* Fisher's exact test.

#### **Table 2.**

PAH levels in wristbands (ng/g), and group differences between mining and control samples, corrected for band days worn.  $N = 101$ .



\* Two-tailed unpaired t-test between mining and control with Satterthwaite correction for unequal variances.

#### **Table 3.**

Multiple linear regression coefficients for association between mining group and PAH levels, controlling for covariates\* .



\* Controlling for sex, age, smoking, education, use of coal in the home and summer season.

#### **Table 4.**

Mean (SD) PAH concentrations in pg/m<sup>3</sup> in outdoor air samples (N = 16), corrected for sampling volume.



\* Based on linear multiple regression model controlling for season.

#### **Table 5.**

Pearson bivariate correlations between wristbands and outdoor air, overall and by group.



 $p^*$  = 0.05.