Study of Human Exposure to Particulate PAHs Using Personal Air Samplers

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Abstract. Three common sources of environmental exposure to particulate polycyclic aromatic hydrocarbons (PAHs) in Taiwan were chosen for this study. They are smoke of incense burning, exhausts of motor vehicles, and fumes of charcoal burning. The campus environment without any specific PAH sources (nonpoint sources) was chosen as the control. The particulate PAH concentrations in the air samples containing smoke of incense burning were only slightly higher than those in the control. However, the total concentration of particulate PAHs in the air samples with exhausts of motor vehicles and fumes of charcoal burning was about 7.5 times and 22 times higher than those observed in the control, respectively. The mean inhalation amounts of particulate PAHs per unit time are very high both in samples with exhausts of motor vehicles (13.9 ng/min) and fumes of charcoal burning (38.1 ng/min). The exposure dose of 22 PAHs per day ranged from 3.18 to 18.0 μg/day under four exposure conditions. Moreover, the personal inhalation BaP$_{eq}$ levels are in the range of 0.4 to 1.55 μg/day.

Polycyclic aromatic hydrocarbons (PAHs) are widely distributed in the environment. Some are considered to be potent carcinogens and mutagens, and hence their fates in the atmosphere and possible effects on humans have been the focus of much attention (Baek et al. 1991; Tsai et al. 2001). In addition, our previous study has suggested that the amount of BghiP is positively correlated with the mutagenicity of airborne particulate samples from Taichung City (Kuo et al. 1998). Recently we have been focusing on some population groups in Taiwan who are regularly exposed to high concentrations of these genotoxic compounds. These groups include motorcyclists, housekeepers who often burn incense indoors, and people broiling meat outdoors with charcoal.

According to a report from the Taiwan Provincial Government Department of Transportation (1999), there are about 500,000 motorcycles and 800,000 automobiles in Taichung City. Because of a large number of vehicles running in this area, large amounts of genotoxic compounds emitted from motor vehicles may have adverse effects on the health of motorcyclists. Most people in Taiwan are Buddhists or Taoists, and they ritually burn incense in their homes. Recently, Chen and Lee (1996) pointed out the genotoxicity and DNA adducts formation of incense smoke condensates. Housekeepers in Taiwan generally spend more than 80% of their time indoors. Long-term exposure to incense smoke possibly makes housekeepers a high-risk group of lung cancer. The third source of PAH exposure is the fumes emitted from charcoal burning at cookouts. In Taiwan, a cookout party is a popular outdoor leisure activity. There are a large number of families who use charcoal to broil food in their back yards or in city parks on holidays, especially during the mid-autumn festival, which is held each year. Numerous cookout parties during this festival produce large amounts of genotoxic compounds within a few days, and participants at these cookouts may inhale dangerous amounts of genotoxic compounds from the cookout fumes.

The purposes of this study are to compare the inhalation amounts of these three sources of air pollution by the respective groups and to estimate the exposure doses of PAHs under four exposure conditions in the real life of Taichung residents. In addition, by collecting samples from the campus (control group), this study also investigates the doses of PAHs to which students are exposed during their normal campus life.

Materials and Methods

Sample Collection

For comparison, all samples were collected with personal samplers (SKC Model 224-PCXR8). Personal environmental monitors (PEM, SKC Model 200) to collect air particles smaller than the cut size (10 μm aerodynamic diameter) were connected to the personal samplers. The personal samplers were operated at a flow rate of 2 L/min with quartz filters (SKC high-purity quartz filter, 37 mm, binder-free). To keep particles from bouncing off the impaction plate, silicon oil was sprayed onto this surface before sampling. A PEM was hanging on the collar of each participant during sampling. The concentration of particles were conditioned at constant temperatures (23 ± 3°C) and at constant humidity (45 ± 5%). The concentrations of particles were obtained by weighing the filter (Mettler AT261, ± 0.01 mg) before and after a run. Before weighing the filters were passed through a Staticmaster (VWR Model 2U500) to eliminate the static electricity. The methods for collecting samples from the four environments are discussed separately in the following.
Exposure to Smoke of Incense Burning

Four homes, in which every family burned incense indoors and no one smoked, were chosen as sampling sites. In each home, four residents each carried one personal sampler and spent most their time in the living room near the shrine (about 1.0 m to 2.0 m). These personal samplers were operated from 5 P.M. to 11 P.M., which is the time of day that most families burn incense in Taiwan. For each home, four samples from four residents were collected simultaneously during the daily sampling time for 5 days. After collecting samples from each family, four samples of 5 days were pooled and analyzed as one composite sample. Thus there were four composite samples from this sampling environment.

Exposure to Exhausts of Motor Vehicles

Samples were collected from street air by five separate motorcycles cruising through Taichung City. On each cruise, five people made up a team, and each rode on one motorcycle separately but cruised together along the same route. Each participant carried a personal sampler. The sampling time for each ride was about 1 h, and the route was changed for each cruise. This kind of ride was taken five times. After collection samples from each cruise were pooled and analyzed as a single sample. Thus there were five samples to be analyzed, and each sample represented a separate cruise (or route).

Exposure to Fumes of Charcoal Broiling

Four different cookouts were selected for sampling in this study. Generally, there were about 8–10 people around a charcoal grill during a cookout. Before cooking, smoky charcoal was put on the ground first and bricks were laid around it. Then meat and food were broiled on the wire netting, which was placed on top of the bricks. Five people (not smoking) were chosen to carry a personal sampler each during the four separate cookouts. Usually cookouts last for 2–3 h. The sampling periods were timed accordingly. After each cookout, the five samples taken at that cookout were pooled and analyzed as a single sample. Thus there were four samples; each comprised the combined samples taken at a separate cookout.

Exposure to the Air Without Specific PAH Sources

Five students were made up a group, and each student carried a personal sampler. Samples were collected on campus with the aim of obtaining data representing the exposure of students to air without specific PAH sources during their normal campus life. The sampling periods were about nine hours, from 8 A.M. to 5 P.M. The samples collected by five students were pooled and analyzed as a single sample. There were five different groups in this study. Students in each group are different. However, they were all chosen from the Department of Public Health, Chung Shan Medical University. All students were asked to stay on the campus during the sampling period. Smoking was not allowed during sampling.

Extraction and Analysis

The particle-loaded quartz filters were extracted with high-purity cyclohexane for 25 min in an ultrasonic bath. The extract was then purified using a silica gel cleanup technique. Compounds retained on silica gel columns were eluted with 50 ml of 1:3 (v/v) dichloromethane and n-hexane. The eluate was filtered through a 5-mm PTFE membrane and was evaporated to dryness using a rotary evaporator. This extraction procedure is described in detail elsewhere (Gundel et al. 1995). Twenty-two PAHs were quantified by gas chromatography with a flame ionization detector (GC-FID) using a Perkin Elmer Autosystem Gas Chromatograph (Model N611-9000). Compounds monitored in this work were naphthalene (NAP), acenaphthylene (ACPY), acenaphthene (ACP), fluorene (FLU), phenanthrene (PHEN), anthracene (ANTHR), fluoranthene (FLA), pyrene (PYR), benzo(c)phenanthrene (BcPH), benzo(b)naphth(2,1-d)thiophene (BNT), cyclopenta(cd)pyrene (CPP), benzo(a)anthracene (BaA), chrysene (CHR), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(e)pyrene (BeP), benzo(a)pyrene (BaP), indeno(1,2,3-cd)pyrene (INP), dibenz(a)anthracene (DBA), benzo(ghi)perylene (BghiP), anthanthrene (ANAN), and coronene(COR). A standard solution consisting of known amounts of 22 PAHs dissolved in dichloromethane was prepared and spiked with a known volume of phenanthrene-d_{10} as the internal standard. The internal standard was also added to the filter samples and processed through the extraction and isolation procedure. Known volumes of PAH standards were spiked into filters to determine the efficiency of the extraction and analysis. This extraction efficiency gave agreements between 80–120% except for NAP, ACPY, and ACP. The lower recoveries (50–60%) that were seen for the lower-molecular-weight species indicated that these species are easy to lose via volatilization during sample preparation. Data reported in this study were not corrected for recoveries. Duplicate samples were within 20% of the sample concentration for all PAHs.

Statistical Methods

Data were analyzed with the statistical package of SPSS 801.C. The Kruskal-Wallis tests were applied to compare the difference of 22 PAHs in each sample from 4 selected environments. Moreover, factor analysis was performed with the principal component method and eigenvalue—one criterion to determine the minimum number of factors to retain. After extraction, varimax rotation was used to improve the interpretability and scientific solution.

Results

The PM\textsubscript{10} concentrations for samples from four environments are shown in Table 1. The data of four different environments showed a significant difference in PM\textsubscript{10} concentration (p = 0.001) when data were performed with a Kruskal-Wallis test. The average PM\textsubscript{10} exposure concentrations for smoke of incense burning and exhausts of motor vehicles were approximately 1.6 and 8.2 times higher, respectively, than the mean value for the control samples taken from normal campus life. However, compared with the control samples, the concentrations of the cookout samples are approximately 62 times higher. The total concentrations of particulate PAHs are also shown in Table 1. Kruskal-Wallis tests show that the total concentrations of particulate PAHs differ significantly (p = 0.002) among the four kinds of environment. The mean concentration of particulate PAHs found at sites with smoke of incense burning is about 1.2 times higher than that for campus life. The sampling condition in this study was somewhat different from those in other studies. For instance, Chen and Lee (1996) sampled smoke condensation from incense burning in a closed room. Because very few genotoxic compounds are emitted from incense burning, it was necessary to burn many sticks