Emission of Polycyclic Aromatic Hydrocarbons and Their Carcinogenic Potencies from Cooking Sources to the Urban Atmosphere

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Traffic has long been recognized as the major contributor to polycyclic aromatic hydrocarbon (PAH) concentrations. However, this does not consider the contribution of cooking sources of PAHs. This study set out, first, to assess the characteristics of PAHs and their corresponding benzo[a]pyrene equivalent (B[a]P$_{eq}$) emissions from cooking sources to the urban atmosphere. To illustrate the importance of cooking sources, PAH emissions from traffic sources were then calculated and compared. The entire study was conducted on a city located in southern Taiwan. PAH samples were collected from the exhaust stacks of four types of restaurant: Chinese, Western, fast food, and Japanese. For total PAHs, results show that the fractions of gaseous PAHs (range, 75.9–89.9%) were consistently higher than the fractions of particulate PAHs (range, 10.1–24.1%) in emissions from the four types of restaurant. But for total B[a]P$_{eq}$, we found that the contributions of gaseous PAHs (range, 15.7–21.9%) were consistently lower than the contributions of particulate PAHs (range, 78.1–84.3%). For emission rates of both total PAHs and total B[a]P$_{eq}$, a consistent trend was found for the four types of restaurant: Chinese (2,038 and 154 kg/year, respectively) > Western (258 and 20.4 kg/year, respectively) > fast food (31.4 and 0.104 kg/year, respectively) > Japanese (5.11 and 0.014 kg/year, respectively). By directly adapting the emission data obtained from Chinese restaurants, we found that emission rates on total PAHs and total B[a]P$_{eq}$ for home kitchen sources were 6,639 and 501 kg/year, respectively. By combining both restaurant sources and home kitchen sources, this study yielded emission rates of total PAHs and total B[a]P$_{eq}$ from cooking sources of the studied city of 8,973 and 675 kg/year, respectively. Compared with PAH emissions from traffic sources in the same city, we found that although the emission rates of total PAHs for cooking sources were significantly less than those for traffic sources (13,500 kg/year), the emission rates of total B[a]P$_{eq}$ for cooking sources were much higher than those for traffic sources (61.4 kg/year). The above results clearly indicate that although cooking sources are less important than traffic sources in contributing to total PAH emissions, PAH emissions from cooking sources might cause much more serious problems than traffic sources, from the perspective of carcinogenic potency. *Key words:* benzo[a]pyrene equivalent concentration, cooking sources, polycyclic aromatic hydrocarbons, traffic sources. *Environ Health Perspect* 111:483–487 (2003). doi:10.1289/ehp.5518 available via *http://dx.doi.org/* [Online 30 October 2002]

Polycyclic aromatic hydrocarbons (PAHs) are one of the first identified airborne carcinogenic pollutants containing two or more aromatic rings that are fused together in different arrangements (*1*). PAHs and derivatives are associated with the incomplete combustion of organic material arising partly from natural combustion such as volcano eruptions or forest fires, but most emissions arise from anthropogenic activities such as the burning of gasoline in motor vehicles, residential heating, home cooking, and industrial production activities (*1*). In the past 30 years, many studies have suggested increased risk for certain cancers in cooks and other food-service workers (*2–7*). Because of this, many researchers have emphasized investigating PAH compositions in indoor air resulting from cooking processes. For example, Rogge et al. (*8*) found that the use of natural gas for cooking would increase the PAH concentrations in indoor air. Siegmann and Sattler (*9*) found that PAH concentrations contained in hot cooking oil fumes (range, 1.08–22.8 μg/m^3) were higher than those in an office room where 96 cigarettes were consumed within 6 hr (1.2 μg/m^3). In

particular, van Houdt et al. (*10*) suggested that the cooking process was the most important contributor to the total mutagenic activity of indoor air. However, canopy hood ventilation has been widely used for cooking sources in many urban areas. Therefore, it can be expected that most PAHs emitted from cooking sources could be exhausted to the urban atmosphere. Many researchers have suggested that traffic is the major contributor to PAH concentrations in the atmosphere of urban and suburban areas (*1*). In one study, Harrison et al. (*11*) indicated that road traffic accounted for 88% of ambient benzo[a]pyrene at an urban location in Birmingham, United Kingdom. But to our knowledge, this estimate does not consider the contribution of cooking sources and hence warrants further investigation.

In this study we first focused on investigating the contents of PAHs that were emitted from stacks of four types of restaurants: Chinese, Western, fast food, and Japanese. Then, PAH emissions from home kitchen sources were estimated according to emission data obtained from Chinese restaurants. In addition, several PAH compounds have been

classified by the International Agency for Research on Cancer (*12*) as "probable" human carcinogens (2A) or "possible" human carcinogens (2B). Therefore, the carcinogenic potency associated with PAH emissions from various cooking sources were also estimated. In this study, we assumed that PAH emissions from both restaurants and home kitchens represented those emitted from all cooking sources. To assess the effect of cooking sources on PAHs emitted into the urban atmosphere, we compared the above-estimated PAH emissions with those emitted from traffic sources in the same city by directly using the emission data presented in our previous studies (*13–15*).

Materials and Methods

Sampling strategy. In this study, a city (area, 2,016 km^2; population, 1,104,682) located in southern Taiwan was selected. According to the statistical data provided by the city government, the city contained 862 restaurants, including 743 Chinese, 88 Western, 20 fast food, and 11 Japanese. However, because of both cost and manpower, only 10 restaurants (4 Chinese, 2 Western, 2 fast food, and 2 Japanese) were randomly selected for this study. Table 1 lists the main cooking methods, types of food oil used, mean food oil consumption rates and cooking time for the total serving of lunch (or dinner), the stack diameters, and the mean stack outlet velocities and temperatures.

During the cooking period for lunch on the sampling day, we collected three PAH samples from the stack of each selected restaurant. We used a PAH sampling system (PSS; Li-Teh Co., Kaoushing, Taiwan) comparable to that specified by modified method 5 (*16*) for sampling semivolatile organic material. This system also has been widely used for collecting PAH samples from various industrial stacks (*17–20*). The sampling system was equipped with a sampling probe, cooling device, glass cartridge, pump, flow meter, and control computer. Each PAH sample was collected isokinetically from the stack, with a

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sampling flow rate of approximately 10 L/min for 45 min per sample. Using a critical orifice flow calibrator (model GMW-25; General Metal Work, Taichung, Taiwan), we determined the accurate sampling flow rate by averaging the flow rates measured at the beginning and at the end of the sampling period. PAHs collected by a tube-type glass fiber filter (25 × 90 mm, Whatman glass fiber thimble) in the sampling probe (i.e., particulate PAHs) were stored in a prebaked glass bottle (wrapped with aluminum foil) for shipment before the chemical analysis. Gaseous PAHs collected by the glass cartridge, packed with a 5-cm polyurethane foam (PUF) plug, followed by a 2.5-cm XAD-16 resin supported by a 2.5-cm PUF plug, were stored in a clean screw-capped jar (with a Teflon cap liner) for transportation. Three breakthrough tests were investigated by using a two-layer XAD-16 cartridge with the sequence in the cartridge as PUF-1, XAD-16–1, PUF-2, XAD-16–2, and PUF-3 (Li-Tex Co., Kaoushing, Taiwan). No significant amounts of PAHs were found in the sections of PUF-2, XAD-16–2, or PUF-3.

PAH analysis. For PAH analysis, each collected sample (including particulate and gaseous PAH samples) was extracted in a Soxhlet extractor with a mixed solvent (*n*-hexane and dichloromethane; vol/vol, 1:1; 500 mL each) for 24 hr. The extract was then concentrated, cleaned up, and reconcentrated to exactly 1.0 or 0.5 mL. PAH contents were determined with a Hewlett-Packard (HP) gas chromatograph (GC) (HP 5890A; Hewlett-Packard, Wilmington, DE, USA) with a mass selective detector (MSD) (HP 59H72) and a computer workstation (Aspire C500;

Acer, Taipei, Taiwan). This GC/MSD was equipped with a capillary column (HP Ultra 2, 50 m × 0.32 mm × 0.17 μm) and an automatic sampler (HP-7673A) and operated under the following conditions: injection volume of 1 μL, splitless injection at 310°C, ion source temperature at 310°C, oven from 50 to 100°C at 20°C/min; 100 to 290°C at 3°C/min; hold at 290°C for 40 min. The masses of primary and secondary ions of PAHs were determined using the scan mode for pure PAH standards. PAHs were qualified using the selected ion monitoring (SIM) mode (*21*).

The concentrations of 21 PAH species were determined: including naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, cyclopenta[*c,d*]pyrene, benzo[*a*]anthracene, chrysene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*e*]pyrene, (B[*a*]P), perylene, indeno[1,2,3-*cd*]pyrene, dibenzo[*a,h*]anthracene, benzo[*b*]chrysene, benzo[*ghi*]perylene, and coronene. The GC/MSD was calibrated with a diluted standard solution of 16 PAH compounds (PAH mixture-610M; Supelco, Bellefonte, PA, USA) plus five additional individual PAHs obtained from Merck (Darmstadt, Germany). Analysis of serial dilutions of PAH standards showed the limit of detection of GC/MSD to be between 0.021 and 0.384 ng for the 21 PAH compounds. The limit of quantification (LOQ) was defined as the limit of detection divided by the sampling volume. The LOQs of the 21 PAH compounds for PSS samples were between 0.047 and 0.853 ng/m^3. Ten consecutive injections of a PAH 610-M standard yielded an average relative standard deviation of

GC/MSD integration area of 3.0%, with a range of 0.8–5.1%. Following the same experimental procedures used for the treatment of samples, we determined recovery efficiencies by processing a solution containing known PAH concentrations. This study showed the recovery efficiencies for the 21 PAH compounds to range from 0.765 to 1.060, with an average value of 0.863. Analyses of field blanks, including the aluminum foil, polyethylene (PE) bottle, glass fiber filter, and PUF/XAD-16 cartridge, revealed no significant contamination (GC/MSD integrated area < detection limit).

Data analysis. In this study, the total PAH concentration was regarded as the sum of the concentrations of 21 PAH compounds for each collected sample. To assess PAH homolog distribution for each collected sample, we further classified total PAHs into three categories: low molecular weights (LM-PAHs, containing two- to three-ringed PAHs), middle molecular weights (MM-PAHs, containing four-ringed PAHs), and high molecular weights (HM-PAHs, containing five- to seven-ringed PAHs). Moreover, considering that several PAH compounds are known human carcinogens, the carcinogenic potencies associated with PAH emissions from each emission source were also determined. In principle, the carcinogenic potency of a given PAH compound can be assessed on the basis of its benzo[*a*]pyrene equivalent concentration (B[*a*]P$_{eq}$). Calculation of the B[*a*]P$_{eq}$ concentration for a given PAH compound requires the use of its toxic equivalent factor (TEF), which represents the relative carcinogenic potency of the given PAH compound, using benzo[*a*]pyrene as a reference compound to adjust its original concentration. Only a few

Table 1. Background information for the four types of restaurant studied.

Background information	Type of restaurant			
	Chinese (*n* = 4)	Western (*n* = 2)	Fast food (*n* = 2)	Japanese (*n* = 2)
Main cooking methods	Stir fry Simmer Steam Roast Smoke Stew	Grill Broil Roast Deep fry Smoke Stew	Deep fry Stew	Steam Simmer Stir fry Stew
Types of cooking oil	Soy bean oil Peanut oil	Soy bean oil Corn oil	Vegetable oil Butter Corn oil	Soy bean oil Corn oil Peanut oil
Cooking time (minutes per lunch or minutes per dinner)				
Mean	145	145	109	155
Range	140–150	140–150	97–121	150–160
Food oil consumption rate (liters per lunch or liters per dinner)				
Mean	13.4	15.5	13.4	16.4
Range	8.80–18.4	14.1–16.8	11.2–16.3	14.3–18.5
Stack outlet velocity (meters per second)				
Mean	9.96	9.48	9.95	14.1
Range	6.21–14.9	6.36–12.6	9.46–10.5	5.79–22.4
Stack diameter (m)				
Mean	0.79	0.90	0.82	0.50
Range	0.47–1.10	0.53–1.27	0.74–0.89	0.35–0.64
Stack outlet temperature (K)				
Mean	315	314	317	314
Range	314–317	312–316	316–317	313–315

Table 2. PAH compounds and their TEFs.[a]

PAH	TEF
Naphthalene	0.001
Acenaphthylene	0.001
Acenaphthene	0.001
Fluorene	0.001
Phenanthrene	0.001
Anthracene	0.01
Fluoranthene	0.001
Pyrene	0.001
Cyclopenta[*c,d*]pyrene	—[b]
Benzo[*a*]anthracene	0.1
Chrysene	0.01
Benzo[*b*]fluoranthene	0.1
Benzo(*k*]fluoranthene	0.1
Benzo[*e*]pyrene	—[b]
Benzo[*a*]pyrene	1
Perylene	—[b]
Indeno[1,2,3-*cd*]pyrene	0.1
Dibenzo[*a,h*]anthracene	1
Benzo[*b*]chrysene	—[b]
Benzo[*ghi*]perylene	0.01
Coronene	—[b]

[a]Data from Nisbet and Lagoy (*24*). [b]No TEF has been suggested.

proposals for TEFs are available (*22–25*). Among them, the list of TEFs completed by Nisbet and LaGoy in 1992 (*25*) (Table 2) has been suggested by Petry et al. (*26*) because it reflects well the actual knowledge of the toxic potency of each individual PAH compound. On the basis of this TEF list, the carcinogenic potency of total PAHs (i.e., total B[*a*]P$_{eq}$) can be assessed by the sum of the B[*a*]P$_{eq}$ concentrations estimated for each PAH compound with a TEF in the total PAHs.

Results and Discussion

Characteristics of PAHs emitted from restaurant sources. Table 3 shows both the means and the ranges of total PAH, LM-PAH, MM-PAH, HM-PAH, and total B[*a*]P$_{eq}$ concentrations (gaseous + particulate phases) of that contained in the stack flue gas of 10 restaurants: 4 Chinese, 2 Western, 2 fast food, and 2 Japanese. The magnitudes of total PAH concentrations for the four types of restaurant

were Western (92.9 μg/m^3) > Chinese (80.1 μg/m^3) > fast food (63.3 μg/m^3) > Japanese (55.5 μg/m^3). Table 3 also shows that the magnitudes of PAH homologs for the above four types of restaurant shared the same trend: LM-PAHs (range 51.5–76.1 μg/m^3) > HM-PAHs (range 3.06–17.6 μg/m^3) > MM-PAHs (range 0.975–3.47 μg/m^3). Based on this, it is not so surprising that the trend for total B[*a*]P$_{eq}$ concentrations for the four types of restaurant was similar to that for total PAHs: Western (4.86 μg/m^3) > Chinese (4.07 μg/m^3) > fast food (0.600 μg/m^3) > Japanese (0.486 μg/m^3). Figure 1 shows the distributions of gaseous PAHs and particulate PAHs contained in total PAHs, LM-PAHs, MM-PAHs, HM-PAHs, and total B[*a*]P$_{eq}$ for the four types of restaurant. For total PAHs, we found that the fractions of gaseous PAHs in the four types of restaurant (range, 75.9–89.9%) were consistently higher than the fractions of particulate PAHs (range, 10.1–24.1%). The above results

suggest that control of gaseous PAH emissions would be more important than control of particulate PAH emissions for the restaurant sources. However, when we examined the contributions of gaseous PAHs and particulate PAHs to total B[*a*]P$_{eq}$ concentrations for the four types of restaurant, we found that the contributions of gaseous PAHs (range 15.7–21.9%) were consistently less than the contributions of particulate PAHs (range 78.1–84.3%). The above results clearly suggest that from the perspective of carcinogenic potency the control of particulate PAH emissions would be more important than the control of gaseous PAH emissions for restaurant sources. Here, the fractions of gaseous PAHs were consistently higher in total PAHs but were consistently lower in total B[*a*]P$_{eq}$ than particulate PAHs in the four types of restaurant; this warrants further discussion. As shown in Figure 1, the fractions of gaseous PAHs in both LM-PAHs and MM-PAHs for the four

Table 3. Mean PAH concentrations (gaseous phase + particulate phase; micrograms per cubic meter) emitted from the stacks of 10 restaurants: 4 Chinese, 2 Western, 2 fast food, and 2 Japanese.

PAH	Chinese		Western		Fast food		Japanese	
	Range	Mean	Range	Mean	Range	Mean	Range	Mean
Naphthalene	15.9–56.2	36.1	39.4–83.2	61.3	22.2–71.3	46.8	14.2–65.8	39.5
Acenaphthylene	1.73–21.6	11.7	1.06–7.36	4.21	1.52–1.58	1.55	0.562–1.94	1.25
Acenaphthene	0.967–2.33	1.65	0.778–1.00	0.890	0.282–0.434	0.360	0.120–0.451	0.285
Fluorene	0.982–3.56	2.32	1.31–1.56	1.44	0.823–3.02	1.92	1.74–3.02	2.38
Phenanthrene	1.96–10.8	6.38	5.71–8.58	7.15	4.89–6.35	5.62	6.35–8.46	7.41
Anthracene	0.474–1.58	1.03	0.454–1.85	1.15	0.189–0.261	0.225	0.191–0.266	0.225
Fluoranthene	0.821–1.83	1.32	0.655–2.05	1.35	0.542–0.901	0.720	0.372–0.908	0.635
Pyrene	1.01–1.65	1.33	0.711–2.47	1.59	0.487–0.492	0.485	0.172–0.483	0.325
Cyclopenta[*c,d*]pyrene	0.232–2.59	1.41	0.054–0.75	0.398	0.911–2.40	1.65	ND–2.24	1.120
Benzo[*a*]anthracene	0.111–1.52	0.814	0.011–0.29	0.150	0.163–0.392	0.276	0.011–0.021	0.015
Chrysene	0.134–0.870	0.502	0.020–0.48	0.247	0.484–0.720	0.596	0.067–0.142	0.104
Benzo[*b*]fluoranthene	0.121–1.63	0.873	0.905–1.49	1.20	0.504–1.00	0.752	ND–0.805	0.403
Benzo[*k*]fluoranthene	0.130–1.50	0.814	0.794–1.78	1.29	0.444–0.621	0.528	ND–0.705	0.352
Benzo[*e*]pyrene	0.245–1.71	0.977	0.389–3.21	1.80	0.143–0.367	0.256	0.051–0.654	0.351
Benzo[*a*]pyrene	0.622–1.82	1.22	0.036–3.15	1.59	0.210–0.393	0.296	0.139–0.420	0.280
Perylene	0.508–2.88	1.69	0.024–2.95	1.48	0.025–0.226	0.216	0.201–0.261	0.232
Indeno[1,2,3-*cd*]pyrene	0.729–4.84	2.81	ND–2.15	1.08	0.134–0.226	0.177	0.542–0.930	0.736
Dibenzo[*a,h*]anthracene	1.26–2.56	1.91	ND–2.70	1.35	0.051–0.096	0.069	ND	ND
Benzo[*b*]chrycene	0.610–5.36	2.98	ND–4.28	2.14	0.094–0.145	0.117	ND–0.322	0.161
Benzo[*ghi*]perylene	0.560–2.73	1.65	0.013–1.29	0.648	0.093–0.233	0.160	ND–0.303	0.152
Coronene	0.399–1.08	0.737	0.033–0.87	0.448	0.281–0.732	0.504	0.242–0.558	0.400
Total PAHs	29.5–130	80.1	35.1–116	92.9	34.5–91.9	63.3	24.7–86.4	55.5
LM-PAHs	21.8–91.6	59.1	40.1–103	76.1	29.9–83.0	56.5	23.2–79.8	51.5
MM-PAHs	1.94–4.99	3.47	1.37–4.80	3.08	1.18–1.78	1.48	0.550–1.40	0.975
HM-PAHs	5.59–29.6	17.6	2.23–25.2	13.7	3.51–7.12	5.32	0.958–5.15	3.06
Total B[*a*]P$_{eq}$	2.95–5.20	4.07	3.01–6.70	4.86	0.484–0.715	0.600	0.314–0.598	0.486

ND, not detectable.
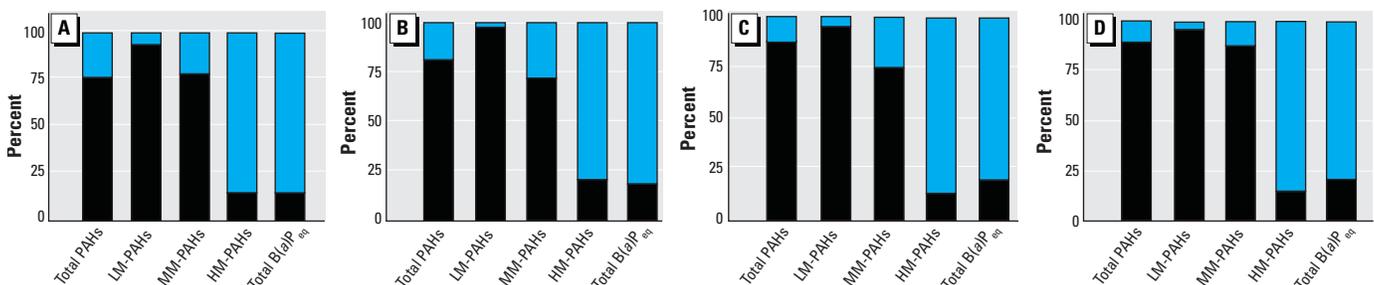
Figure 1. Distributions of gaseous PAHs and particulate PAHs contained in total PAHs, LM-PAHs, MM-PAHs, HM-PAHs, and total B[*a*]P$_{eq}$ for the four types of restaurant: (*A*) Chinese, (*B*) Western; (*C*) fast food, (*D*) Japanese.

types of restaurant (ranges, 93.9–97.5% and 72.3–87.7%, respectively) were higher than the fractions of particulate PAHs (range 2.46–6.11%). But the fractions of gaseous PAHs in HM-PAHs (range 13.7–21.0%) were much lower than those of particulate PAHs (range 79.0–86.3%). Considering that total PAHs contained in the stack flue gas of the four types of restaurant were composed mainly of LM-PAHs (Table 3), it is not so surprising that gaseous PAHs accounted for higher fractions in total PAHs. Conversely, it is known that PAHs with higher molecular weights are associated with the higher TEFs (Table 2). In addition, it should be noted that particulate PAHs were mainly contributed by the HM-PAHs. Based on these, it is not surprising that particulate PAHs accounted for higher factions in total $B[a]P_{eq}$ than did gaseous PAHs in the four types of restaurant.

PAH emission factors for restaurant sources. It is known that PAH samples collected from the four types of restaurants were associated with different cooking methods, food oil consumption rates, cooking time, stack outlet velocities, and stack diameters (Table 1). Therefore, using the total PAH and total $B[a]P_{eq}$ concentrations might not properly reflect their emission intensities. For this, PAH emission factors on both total PAHs and total $B[a]P_{eq}$ (denoted EF_{totPAH} and $EF_{totB[a]Peq}$, respectively) for the four types of restaurant were calculated to compare their emission intensities. Here, EF_{totPAH} and $EF_{totB[a]Peq}$ (mg/L food oil) were calculated, respectively, according to the following two equations:

$$EF_{totPAH} = (\text{total PAH concentration}) \times [(1/4) \times \pi \times d^2] \times v \times 60 \times t \times 10^{-3}/CR_{food\text{-}oil}$$

$$EF_{totB[a]Peq} = (\text{total } B[a]P_{eq} \text{ concentration}) \times [(1/4) \times \pi \times d^2] \times v \times 60 \times t \times 10^{-3}/CR_{food\text{-}oil},$$

where d, v, t, and $CR_{food\text{-}oil}$ are, respectively, the stack diameter (in meters), stack outlet velocity (in meters per second), cooking time (in minutes per lunch or minutes per dinner), and food oil consumption rate (in liters of food oil per lunch or liters of food oil per dinner) (Table 1). As shown in sequence for the magnitudes of EF_{totPAH} for the four types of restaurant, we found Chinese (281 mg/L food oil) > Western (259 mg/L food oil) > fast food (156 mg/L food oil) > Japanese (37.8 mg/L food oil) (Table 4). A similar trend can also be found for $EF_{totB[a]Peq}$ for the four types of restaurant (21.2, 20.5, 0.518, and 0.106 mg/L food oil for Western, Chinese, fast food, and Japanese, respectively) (Table 4). These results indicate that both Chinese and Western restaurants were comparable on EF_{totPAH} and $EF_{totB[a]Peq}$. However, it should be noted that the mean

EF_{totPAH} for Chinese restaurants was 1.80- and 7.43-fold higher than those for fast-food and Japanese restaurants, respectively. However, the mean $EF_{totB[a]Peq}$ for Chinese restaurants was 40.9- and 200-fold in magnitude higher than those for fast-food restaurants and Japanese restaurants, respectively. These results suggest that PAH emissions from both fast-food restaurants and Japanese restaurants not only contained lower total PAH contents but also had much lower carcinogenic potencies. At this stage, it is known that both EF_{totPAH} and $EF_{totB[a]Peq}$ can be affected strongly by the cooking method and the type of food oil. Because the mechanisms associated with the formation of PAHs for various cooking sources were not known, this area warrants further investigation.

PAH emission rates for restaurant and home kitchen sources. We assume that both the mean emission factors and food oil consumption rates obtained from this study are representative of the four types of restaurant. In addition, we assumed that all restaurants ran for 365 days per year and served only lunch and dinner each day. Based on these assumptions, the total PAH and total $B[a]P_{eq}$ emission rates (denoted ER_{totPAH} and $ER_{totB[a]Peq}$, respectively, in kilogram per

year) for a given type of restaurant could be determined, respectively, according to the following two equations:

$$ER_{totPAH} = (EF_{totPAH}) \times CR_{food\text{-}oil} \times n \times 2 \times 365 \times 10^{-6},$$

$$ER_{totB[a]Peq} = (EF_{totB[a]Peq}) \times CR_{food\text{-}oil} \times n \times 2 \times 365 \times 10^{-6},$$

where n was the total number of the given types of restaurant in Tainan, Taiwan (743, 88, 20, and 11 for Chinese, Western, fast food, and Japanese, respectively). Results show ER_{totPAH} and $ER_{totB[a]Peq}$ for the four types of restaurant were Chinese (2,038 and 154 kg/year, respectively) > Western (258 and 20.4 kg/year, respectively) > fast food (31.4 and 0.104 kg/year, respectively) > Japanese (5.11 and 0.014 kg/year, respectively) (Table 5).

In addition to restaurants, it is believed that home kitchens might also play an important role in PAH emissions into the urban atmosphere. According to the internal statistics data provided by the Taiwan Food Oil Producer Association, the personal consumption rate of food oil ($PCR_{food\text{-}oil}$) in the Taiwan area was approximately 58.7 mL/person/day. We assumed that both EF_{totPAH} and $EF_{totB[a]Pq}$ data obtained from Chinese

Table 4. Mean emission factors of EF_{totPAH} and $EF_{totB[a]Peq}$ for the studied four types of restaurant (milligrams per liter of food oil).

Emission factor	Chinese ($n = 4$)	Western ($n = 2$)	Fast food ($n = 2$)	Japanese ($n = 2$)
EF_{totPAH}				
Mean	281	259	156	37.8
Range	148–401	150–368	130–182	37.1–40.4
$EF_{totB[a]Peq}$				
Mean	21.2	20.5	0.518	0.106
Range	10.4–40.2	1.53–45.4	0.449–0.588	0.092–0.121

Table 5. Estimated annual emission rates on total PAHs and total $B[a]P_{eq}$ for the cooking source (including restaurant and home kitchen sources) and the traffic source in the studied city area.

Emission source	n	Food oil (or fuel) consumption rate[a]	Emission factor[b] Total PAHs	Emission factor[b] Total $B[a]P_{eq}$	Emission rate[c] Total PAHs	Emission rate[c] Total $B[a]P_{eq}$
Cooking sources						
Types of restaurant						
Chinese	743	13.4	281	21.2	2,038	154
Western	88	15.5	259	20.5	258	20.4
Fast food	20	13.8	156	0.518	31.4	0.104
Japanese	11	16.4	37.8	0.106	5.11	0.014
Total					2,334	174
Home kitchens	1,104,682	58.7	281	21.2	6,639	501
Total					8,973	675
Traffic sources						
Truck/bus	8,672	27.1	37.2	0.127	8,730	29.8
Car	261,291	1.34	5.21	0.063	1,830	22.0
Four-stroke motorcycle	347,780	0.218	13.2	0.081	1,000	6.15
Two-stroke motorcycle	225,196	0.178	49.2	0.086	1,970	3.44
Total					13,500	61.4

[a]Representing the mean food oil consumption rates for restaurant sources (in liters food oil/lunch or dinner/restaurant) and home kitchen sources (in milliliters per person per day), and the fuel consumption rates for traffic sources (in kiloliters per vehicle per year). [b]Representing emission factors for restaurant sources (in milligram per liter of food oil), home kitchen sources (in milligrams per liter of food oil), and traffic sources (in milligrams per liter of fuel). [c]Representing the annual emission rates for restaurant sources, home kitchen sources, and traffic sources (in kilogram per year).

restaurants (281 and 21.2 mg/L food oil, respectively) were representative of those found in home kitchens. Then, both ER_{totPAH} and $ER_{totB[a]Peq}$ for home kitchen (in kilograms per year) can be estimated, respectively, according to the following two equations:

$$ER_{totPAH} = (EF_{totPAH}) \times PCR_{food\text{-}oil} \times 10^{-3} \times n \times 365 \times 10^{-6}$$

$$ER_{totB[a]Peq} = (EF_{totB[a]Peq}) \times PCR_{food\text{-}oil} \times 10^{-3} \times n \times 365 \times 10^{-6}$$

where n was the population of the studied city area (1,104,682, as mentioned above). This study yielded ER_{totPAH} and $ER_{totB[a]Peq}$ of 6,639 and 501 kg/year, respectively, for home kitchens. These values were higher than those for restaurants (i.e., combining the four types of restaurant together equals 2,334 for ER_{totPAH} and 174 kg/year for $ER_{totB[a]Peq}$) (Table 5). Nevertheless, these results also suggest that both home kitchens and restaurants should be considered when estimating PAH emissions from cooking sources.

Comparison of PAH emission rates between cooking and traffic sources. Assuming PAH emissions from cooking sources were equivalent to those from all restaurants and home kitchen sources, this study yielded ER_{totPAH} and $ER_{totB[a]Peq}$ for cooking sources of 8,973 and 675 kg/year, respectively (Table 5). Traffic sources have long been recognized as the major contributor of PAHs in urban areas (*1,11*). Therefore, to assess the importance of cooking sources, PAH emissions from traffic were also estimated. According to the statistics data provided by the Transportation Bureau in Tainan, there were 842,939 motor vehicles in the city comprising 8,672 buses/trucks, 261,291 cars, 347,780 four-stroke motorcycles, and 225,196 two-stroke motorcycles. The consumption rates of fuel (CR_{fuel}) for the four types of motor vehicle listed above were 27.1, 1.34, 0.218, and 0.178 kL/vehicle/year, respectively. For simplicity, we assumed all bus/trucks used diesel and the other three types of motor vehicles used 95 lead-free gasoline. On the basis of our previous findings (*13–15*), EF_{totPAH} and $EF_{totB[a]Peq}$ for the four types of motor vehicle listed above were 37.2 and 0.127 mg/L fuel, 5.21 and 0.063 mg/L fuel, 13.2 and 0.081 mg/L fuel, and 49.2 and 0.086 mg/L fuel, respectively. Based on these, ER_{totPAH} and $ER_{totB[a]Peq}$ (in kilograms per year) for a given traffic source can be estimated, respectively, according to following two equations:

$$ER_{totPAH} = (EF_{totPAH}) \times CR_{fuel} \times 10^3 \times n \times 10^{-6},$$

$$ER_{totB[a]Peq} = (EF_{totBaPeq}) \times CR_{fuel} \times 10^3 \times n \times 10^{-6},$$

where n was the number of vehicles for the given traffic source, as above. Results show the total ER_{totPAH} and $ER_{totB[a]Peq}$ for all traffic sources (by combining buses/trucks, cars, four-stroke motorcycles, and two-stroke motorcycles) were 13,500 and 61.4 kg/year, respectively (Table 5). These results suggest that ER_{totPAH} from cooking sources (8,973 kg/year) was approximately 0.66-fold that from traffic sources, but $ER_{totB[a]Peq}$ from cooking sources (675 kg/year) was approximately 11.0-fold that from traffic sources. These results clearly suggest that in Tainan PAH emissions from cooking sources are much less important than those from traffic sources. However, the carcinogenic potency of PAH emissions from cooking sources was much greater than that from traffic sources.

Conclusions

In all restaurant sources studied, the emissions of gaseous PAHs were greater than those of particulate PAHs. However, the carcinogenic potency of gaseous PAH emissions was less than that of particulate PAH emissions. PAH emission intensities for the four types of restaurants for both EF_{totPAH} and EF_{totPAH} shared the same trend: Chinese > Western > fast food > Japanese. For cooking sources, we found that both ER_{totPAH} and $ER_{totB[a]Peq}$ from home kitchen sources were consistently higher than those for restaurant sources. Nevertheless, these results suggest both home kitchens and restaurants should be included for estimating PAH emissions from cooking. To determine the significance of cooking sources, PAH emissions from traffic sources were estimated. We found that although ER_{totPAH} from cooking sources was approximately 0.66-fold that from traffic sources, $ER_{totB[a]Peq}$ from cooking sources was approximately 11.0-fold that from traffic sources. These results clearly suggest that, in addition to PAHs emitted from traffic, cooking sources make an important contribution to PAH emissions into the ambient environment of an urban area. However, it should be noted that other city areas might have intrinsic differences in cooking methods, food oil consumption rates, restaurant compositions, and traffic conditions compared with those found in this study. Therefore, the importance of cooking sources as contributors to PAH emissions in other city areas could be somewhat different than that found in this study. Therefore, further investigation is needed.

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