Effects of Fish (Mackerel Pike) Broiling on Polycyclic Aromatic Hydrocarbon Contamination of Suspended Particulate Matter in Indoor Air

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The effects of fish (mackerel pike) broiling on indoor air pollution with polycyclic aromatic hydrocarbons (PAH) were investigated using personal cascade impactors (PCI) sampler and a HPLC spectrofluorometric method. Particles in indoor air were separately collected into 3 classes according to their diameters (≥10 μm, 2.5–10 μm, and ≤2.5 μm). Acetonitrile could extract PAH, even from fatty particulates generated during fish broiling. PAH were mostly contained in the smallest class (≤2.5 μm) particulates. The indoor air was polluted with high levels of PAH during broiling when ventilation was not in operation. The benzo[*a*]pyrene concentration in the smallest particulates at that time was lower at the center of the room than that at the corner opposite to the cooking stand. The PAH concentrations did not always decrease depending on the straight-line distance from the emission source (cooking table). As a result, fish broiling is one of the most important sources of indoor air pollution in Japanese houses.

Key words —— indoor air pollution, polycyclic aromatic hydrocarbons, fish broiling, airborne particle, cascade impactor

INTRODUCTION

A variety of chemical substances are included in environmental air, and some of them show carcinogenicity and/or mutagenicity. Through breathing, we continually uptake these chemical substances into our bodies, and chronic effects of such long-term exposure to these untoward substances are feared. Of these chemical substances, many carcinogens, including polycyclic aromatic hydrocarbons (PAH), are known to be contained in airborne particles (AP), particularly those of relatively small particle diameters.[1–3] The ratio of deposition of such suspended particulates on respiratory organs is closely related to particle diameter, and it is known that the smaller the particle diameter, the higher the deposition ratio on pulmonary alveoli or peripheral bronchitis.[4] Since we spend most of our daily time in rooms, it has become important to learn the real state of carcinogenic/mutagenic substance contamination of indoor air, or the condition of personal exposure to such hazardous elements in rooms, in addition to external atmospheric pollution.

Using personal cascade impactors (PCI) samplers which can collect and precisely classify AP according to their diameters, we collected AP inside/outside rooms of general households, and measured the mutagenicity and PAH concentration of substances extracted from the particles with organic solvents. As a result, it became clear that generally higher PAH concentration and stronger mutagenicity is shown by outdoor air samples rather than indoor air samples,[5] and that higher PAH concentration and stronger mutagenicity is associated with smaller particle diameter.[6–8] Meanwhile, there were cases in which higher PAH concentration and stronger mutagenicity was found in indoor air samples compared with outdoor air samples,[9] and the cause in such cases was suggested to be the influence from

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cooking,[10–12] which is one of the main factors in room air contamination.

In this study, broiling is taken up as the possible main source of contamination of indoor air. Mackerel pike is a kind of fish containing a fairly large amount of fat, likely to produce smoke during cooking, and it is common in general homes in Japan. Therefore, this fish was chosen as the sample for this study, and the smoke generated as a result of its broiling was collected by PCI samplers. The method for measuring the content of PAH in indoor AP that resulted from the smoke was subsequently investigated. Particle distribution according to size, differences in data depending on the monitoring points, and the time course of PAH concentration in indoor air before and after cooking, are also investigated.

MATERIALS AND METHODS

Reagents —— Mackerel pike used for testing were frozen products that were purchased and broiled after thawing. Their weights averaged 163 g per fish. PAH standard reagents were pyrene (Py), benz[*a*]anthracene (BaA), and benzo[*a*]pyrene (BaP), all of which were purchased from Wako Pure Chemicals Industries, Ltd. (Japan). Benzo[*k*]-fluoranthene (BkF), benzo[*b*]chrysene (BbC), and dibenz[*a*,*h*]anthracene (dBahA), all of which were produced by Koch-Light Laboratory, Ltd. (U.K.), benzo[*ghi*]perylene (BghiP) was purchased from Aldrich Chemical Co., Inc. (U.S.A.), and dibenzo[*a*,*e*]pyrene (dBaeP) was produced by Chemicals Procurement Laboratory (U.S.A.). Solvents for extraction: acetonitrile, dichloromethane, benzene, and ethanol (Wako Pure Chemical Industries, Ltd., Japan), were of residual agricultural chemicals grade. The solvent for HPLC: acetonitrile (HPLC grade) was purchased from Kokusan Chemical Works, Ltd. (Japan), and distilled ion-exchange water was also used.

Sample Collection through a High-Volume (HV) Sampler —— A mackerel pike was cut into two equal halves, and they were broiled on a metal net (over a gas range) for about 10 min. Particles generated from this fish were collected on a quartz-fiber filter (Pallflex Products, Co. (U.S.A.), 2500QAT-UP, 8×10 inches; herein referred to as "filter") through a stainless steel hood/heatproof-duct-attached HV sampler (Shibata Scientific Technology, Ltd., Japan, HVS-1000). The post-collection filter was folded with the collection side placed inside, shielded from light with aluminum foil, put in a chuck-attached polyethylene bag, and stored in a freezer (at –80°C) until it was used for extraction.

Collection of Samples through PCI Sampler —— The layout of the room where sample collection was carried out, using PCI sampler (Tokyo Dylec Co., Ltd., Japan; and mini-pumps: MP-603T, produced by Shibata Scientific Technology, Ltd., Japan), is shown in Fig. 1. With the following situations set as model cases: mackerel pike was broiled with and without ventilation and sample collection was carried out.

In the case with a ventilating fan in operation, while the fan was being operated, the cut halves of five mackerel pike were broiled, one after another in succession, on a metal net, and in this process, smoke was generated for 60 min. In this connection, PCI samplers were placed at points A, B, and C, as shown in Fig. 1. Before use, the filter was washed with dichloromethane of residual agricultural chemicals grade, and dried. AP were collected at a flow rate of 3 l/min with 50% cut-off values for each particle diameter of ≥10 μm, 2.5–10 μm, and 2.5 μm ≥ on the filter. Collection was carried out during the periods from 10:45 on the day before cooking to 13:00 on the day of cooking (background), from 15:05 to 16:35 (cooking time was from 15:05 to 16:05), from 17:14 to 19:14, and from 21:25 to 15:05 of the following day. The post-collection filter was stored in the same manner as described above.

In the case with a ventilating fan not operating, the cut halves of four mackerel pikes were broiled and smoke was generated, in the same manner as described above. Meanwhile, PCI samplers were placed at points A to G, as shown in Fig. 1, and particulates were classified and collected on the filter at a flow rate of 3 l/min. The collection was carried out during the periods from 11:07 on the day before cooking to 11:08 on the day of cooking (time-period 1, background), from 11:20 to 13:20 (time-period 2, cooking time was from 11:20 to 12:20), from 13:29 to 15:29 (time-period 3), from 15:35 to 17:35 (time-period 4), from 17:41 to 19:41 (time-period 5), and from 19.52 to 11:00 of the following day (time-period 6). The post-collection filter was stored in the same manner as described above.

Extraction Method —— Samples collected through a HV sampler: Two disks with a diameter of 20 mm (1.5% of the collected area of the filter) and 43.0 mg of particles, were cut off from the post-collection filter using a belt punch, cut to small pieces and put

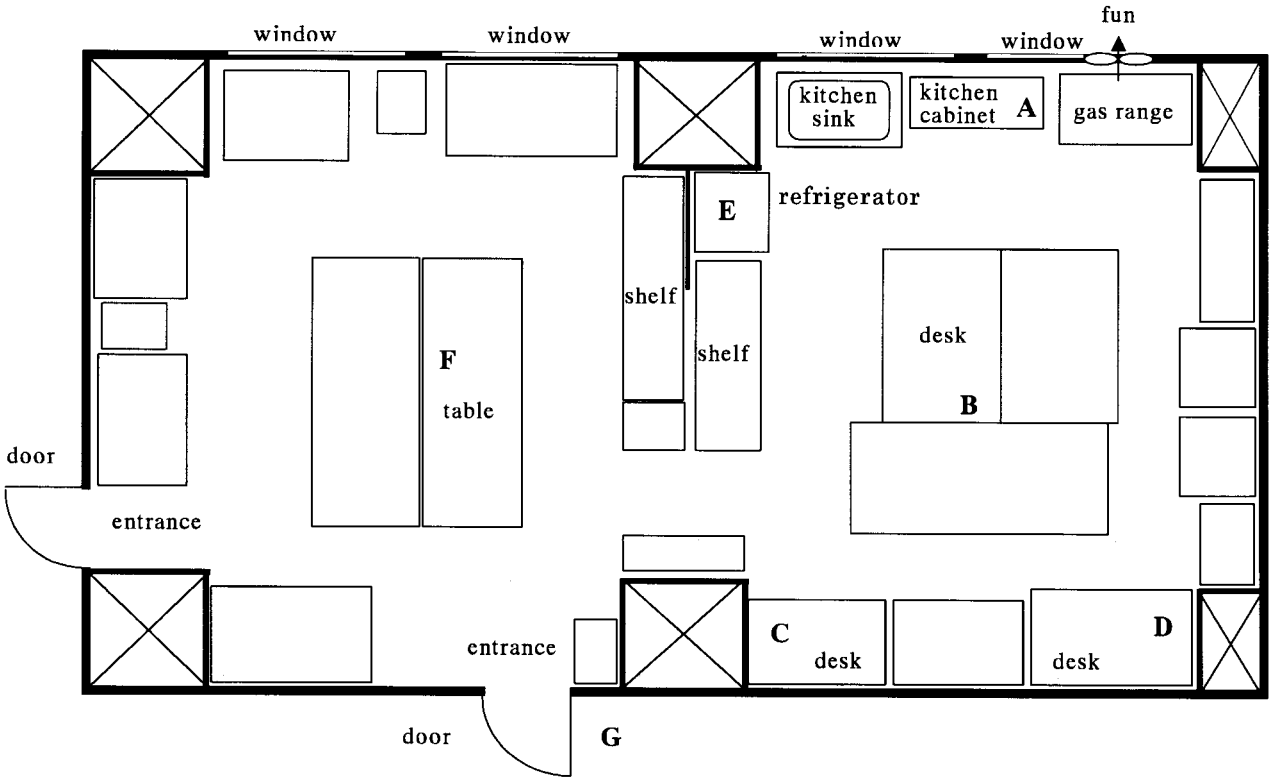

Fig. 1. Layout of the Room and Monitoring Points

The floor space was approximately 152 m^3 (7.6 m × 5.4 m) with a height of 3.7 m. Monitoring point A was 1.1 m distant from the gas range and 2.1 m from the floor, and the other points were approximately 1.5 m from the floor.

in a 8-ml screw-cap test tube. After 6.0 ml of extraction solvent was added, PAH was extracted by sonication two times for 15 min. If the extraction solvent was not acetonitrile, 5.0 ml of extracted solution was evaporated under mild N_2 stream and dissolved in 6.0 ml of acetonitrile. Subsequently, the solution was centrifuged at 3000 rpm for 20 min, and the supernatant solution was used as a sample for PAH analysis.

Samples collected through a PCI sampler: Each post-collection filter was cut to small pieces and put in test tubes, 6.0 ml of acetonitrile was added and extracted in the same manner.

Analysis of PAH —— Analysis subjects were 8 types of PAH: Py, BaA, BaP, BkF, BbC, dBahA, BghiP, and dBaeP. Authentic PAH was dissolved in dimethyl sulfoxide (fluorescence analysis-use, Wako Pure Chemical Industries, Ltd., Japan), diluted with acetonitrile, and used as a standard PAH solution. For PAH analysis, a HPLC apparatus equipping double-columns/fluorescence spectrophotometer[8,13] was used. The principal components of this equipment were as follows: a degasser, ERC-3520 (Erma Optical Works Ltd., Japan); three pumps, LC-6A; a system controller, SCL-6A; an autoinjector, SIL-6A; a column oven, CTO-2A; a fluorescence detector, RF-10AXL (all of these were manufactured by Shimadzu Co., Japan); a mixer, 655-0486 (Hitachi Ltd., Japan); a concentration column, Kaseisorb ODS-60-5 (4.6 mm × 30 mm, Tokyo Chemical Industry Co., Ltd., Japan); and a separation column, Kaseisorb ODS-60-5 (4.6 mm × 250 mm, Tokyo Chemical Industry Co., Ltd., Japan). After application of the sample solution, PAH was concentrated by the first column with a mixed solution of acetonitrile/H_2O = 1 : 1 for 5 min at a flow rate of 1.0 ml/min. Then, the respective PAHs were separated by the second column with acetonitrile /H_2O=8 : 2 at a flow rate of 1.0 ml/min. Table 1 shows the excitation and emission wavelengths used for respective PAH analyses. All of the excitation and emission wavelengths during detection were set and automatically controlled by computers.[13]

RESULTS AND DISCUSSION

Extraction Solvents for PAH

As an extraction solvent for PAH in AP, dichloromethane or a mixture of benzene and etha-

Table 1. Conditions and Determination Limits in PAH Analyses by HPLC

	Wavelength (nm)		Determination limit*
	Excitation	Emission	(ng)
Pyrene	339	373	0.0050
Benz[a]anthracene	292	412	0.0019
Benzo[k]fluoranthene	370	406	0.0061
Benzo[a]pyrene	370	406	0.0024
Dibenz [a,h]anthracene	302	397	0.0029
Benzo[b]chrysene	302	397	0.0026
Benzo[ghi]perylene	370	406	0.0124
Dibenzo[a,e]pyrene	305	398	0.0091

*S/N = 10.

nol has been used. However, these solvents are currently being replaced by acetonitrile and methanol. Since the former two solvents are suspected to carcinogenic and hazardous and use is prohibited in some countries. Therefore, it is difficult to compare the data on samples extracted with respective solvents. Moreover, the former two solvents used for dissolving extracts need to be replaced with other suitable solvents as acetonitrile for HPLC analysis, because dichloromethane and benzene dissolve ODS on the solid phase in the column. When acetonitrile/water is used as the solvent system of HPLC analysis, extraction of PAH with acetonitrile, which can be directly injected into the HPLC in large quantities, is highly advantageous. Meanwhile, in the case of collecting smoke generated from broiling fish with a large fat content, such as mackerel pike, we had some concern about the possible inhibitory effects of fat on the extraction of PAH. Therefore, we investigated the effects of fat in two ways. First, we examined differences in PAH extraction efficiencies with three types of extraction solvent: acetonitrile, a mixture of benzene/ethanol (3 : 1, v/v), and dichloromethane, using an HV sample filter used in the collection of smoke from broiling mackerel pike. PAH were extracted from two disks of the filters using these three solvents. Individual PAHs thus extracted were subsequently analyzed and the results were compared. Consequently, BkF, dBahA, BbC, and dBaeP were less than the determination limit (Table 1), but Py, BaA, BaP, and BghiP could be quantified. Results obtained from five repetitions of independent analyses are shown in Table 2. Although the level of BghiP extracted with acetonitrile was somewhat low, there was little difference among the PAH levels obtained with the three kinds of solvents.

Second, the effect of fat on extraction efficiency using acetonitrile was investigated. Two disks were cut off from the post-collection filter by an HV sampler, and PAH content (the sample value) was measured. Two other disks were cut off from the same filter, 160 μl of PAH standard solution (PAH dissolved in acetonitrile at a concentration of about 375 ng/ml) was added to the disks (80 μl for each disk), and they were dried at room temperature. The same volume of standard solution was added to two disks of control filter, and they were also dried. Further, the same volume of standard solution was added to the test tube, and it was dried up. Each was then extracted by sonication using 6 ml of acetonitrile and analyzed by HPLC. The recovery ratios are shown in Table 3 (recovery level from the standard in the test tube was evaluated as 100%). PAH recovery ratios from the post-collection disks, on which the PAH standard solution was added, were calculated using the increase of each PAH from the sample value. As a result, PAH on the post-collection disks could be satisfactorily recovered by this method. These results suggested that even in samples containing much fat, acetonitrile could be used as an extraction solvent for PAH, as in the case of AP.

Collection Precision of PCI Samplers

The above study was exclusively using an HV sampler, designed to collect particles at a high flow rate without classifying particles. In order to evalu-

Table 2. Extraction of PAH with 3 Solvents from Airborne Particulate Collected by HV Sampler on Filter Disks

PAH	Content of PAH (ng/filter)		
	Acetonitrile	Dichloromethane	Benzene : Ethanol (3 : 1, v/v)
Pyrene	2683 ±160 (5.99%)	2653 ±57 (2.18%)	2625 ±55 (2.11%)
BaA	1382 ± 53 (3.84%)	1416 ±37 (2.58%)	1398 ±41 (2.92%)
BaP	618.2± 39 (6.31%)	651.2± 5.8 (0.90%)	643.5±15.2 (2.36%)
BghiP	867.1± 40.7 (4.70%)	1008 ±58 (5.77%)	974.1±42.8 (4.39%)

PAH were extracted with the respective solvents of acetonitrile, dichloromethane, and a mixture of benzene and ethanol. Number in parentheses reveals the coefficient of variation. $n = 5$.

Table 3. Extraction Efficiencies of PAH which Were Added to a Filter after Collecting Airborne Particulates by a HV Sampler and to a Control Filter

Filter	Extraction ratio of PAH (%)						
	Pyrene	BaA	BkF	BaP	dBahA	BghiP	dBaeP
Control filter	86.01± 2.90	97.31± 3.14	98.73±2.82	98.76±3.16	102.1±3.7	104.4±4.6	104.2±6.1
Sampled filter	84.14±37.24	103.7 ±14.5	103.1 ±3.9	104.8 ±8.0	100.6±5.4	104.8±9.2	115.4±4.5

PAH was extracted with acetonitrile and analyzed by HPLC. PAH contents of a sampled filter were calculated as values increased from the PAH level in airborne particulates, which was quantified beforehand. $n = 5$.

Table 4. Measurement Precision of PAH Concentrations in the Small Particles of Airborne Particulates Collected by Low- Flow, Small-Typecascade Impactors in a Relatively Clean Room

	PAH							
	Pyrene	BaA	BkF	BaP	dBahA	BbC	BghiP	dBaeP
Contents (ng/filter disk[a)])	0.50	0.40	0.33	0.76	0.09	0.10	0.88	0.08
S D[b)]	0.02	0.01	0.01	0.01	0.00	0.00	0.02	0.00
CV[c)] (%)	4.87	1.50	1.80	0.73	3.71	1.30	2.50	5.71

a) diameter of a filter disk was 47 mmϕ. *b*) standard deviation. *c*) coefficient of variation. $n = 4$.

ate precisely the contamination of air in a limited space, such as general homes, it is necessary to use a low-flow sampler that can collect and measure only part of the air without affecting the concentration of the overall air in the room. Furthermore, to carry out a particle diameter-based classified collection, a cascade sampler was used. Meanwhile, the total air volume sucked by a PCI sampler is only about 4 m^3, even collecting for 24 hr, which is much smaller than that by an HV sampler (1000 l/min). Because particles are collected in three different diameter sizes, the sample quantity for each size will be a fairly low level, and therefore collection precision may be affected by factors that are disregarded in the case of an HV sampler. As such, before collecting samples during mackerel pike broiling, with the PCI samplers, their collection precision was checked. Namely, four PCI samplers were set in parallel in the center of a room (point B in Fig. 1), AP were collected at a rate of 3 l/min for 24 hr, and PAH on the filter were extracted with acetonitrile and analyzed by HPLC. Since the quantity of particles collected was so small, precise measurement of PAH concentration was difficult in the large and medium particles, but relatively accurate measurement was possible for the eight kinds of PAH in small particles. Table 4 shows the average PAH concentration in the small particles. The standard deviation (S.D.) and coefficient of variation (C.V.) of the data obtained by four PCI samplers are also shown. For BaP, featuring the highest sensitivity, C.V. was 0.73, while for dBaeP, characterized by the greatest variation, C.V. was 5.71. In view of such results, the collection precision of the PCI sampler regarding the eight kinds of PAH was good, at least for the small particles, so it was judged that there were no problems in practical-use regarding the measurement of PAH in small particles.

Collection of Particles Generated during Mackerel Pike Broiling Using PCI Samplers

Based on the results of the above study, it was deemed possible to examine the effects of cooking (mackerel pike broiling) on indoor air pollution using the PCI sampler. Accordingly, we carried out two kinds of model experiments: broiling mackerel pike on a metal net, and collecting smoke by the PCI samplers, in the cases of with and without ventilation. As a result, the concentrations of BkF, dBahA, BbC, and dBaeP were less than the determination limits for all particle sizes, irrespective of ventilation. BghiP was also below the determination limit in most samples. It was only determinable for the small particles, when the ventilating fan was not operated. Py, BaA, and BaP were determinable in most samples, and their concentrations showed very similar patterns. Table 5 shows the BaP concentration in small particles collected at each monitoring point in Fig. 1 during cooking without ventilation. BaP levels at points verging on the walls were high. The level at each point before cooking was 0.2–0.5 ng/m^3, and it returned to the same level (0.3–0.6 ng/m^3) after 6 hr from the end of the cooking. The smoke was mainly found to go up to the ceiling and spread along

Table 5. BaP Concentration in the Small Particles of Airborne Particulates Collected at 7 Monitoring Points during Fish-Broiling with Ventilation and without Ventilation

Monitoring points[a)]	BaP concentration (ng/m^3)	
	Unventilation[b)]	Ventilation[c)]
A	25.35	1.023
B	12.51	0.654
C	28.21	0.909
D	28.16	*d*)
E	21.63	*d*)
F	21.24	*d*)
G	0.416	*d*)

a) Monitoring points are shown in Fig. 1. *b*) The samplers were operated during broiling for 60 min. *c*) The samplers were operated during broiling for 90 min. *d*) Collection was not performed.

it and then proceed down along the wall. There was a narrow wall at the left side of point E and a beam in the center of the ceiling divided it into right and left sides. The height of the beam was 1.2 m, and an aperture between it and shelves was 50 cm high. A part of the smoke went through this aperture and down to the left side of the room; therefore, the BaP level at point F was also comparatively high.

Figure 2 shows the time course of BaP concentration at the cooking stand (point A), when the ventilating fan was not operated, and outside the room (point G). At the cooking stand, the BaP concentration in the small particles was about 0.5 ng/m^3 before cooking, but it became 25 ng/m^3 during the cooking; *i.e.*, BaP concentration in small particles remarkably increased by fish broiling. Although BaP concentrations in the large and medium particles also increased, the ratio of increase of BaP in the small particles was much higher compared with these larger particles. After cooking, the PAH concentration declined with the time, and by the following day the concentration became almost the same level as that before cooking. Since the BaP level outside the room was almost unchanged through the sampling period, cooking was confirmed to be the main source of PAH contamination in the room. On the other hand, from the time course of BaP concentration in the small particles at point A with ventilation (data not shown), the BaP level was maintained at the highest level (1.02 ng/m^3) at time-period 2 (the cooking time) throughout the experiment. Comparing this value, the level without ventilation was 25-fold higher. This result reflects the concentrations of other PAH, *e.g.*, the concentration of Py was about 20-fold higher, and about 10-fold higher for BaA,

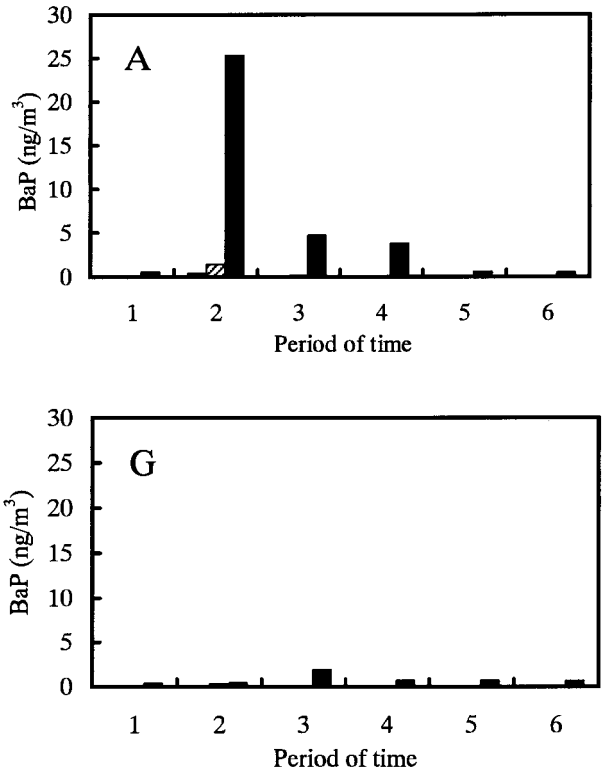

Fig. 2. BaP Concentration Changes in Size of Particles Over Time Inside (A)/Outside (G) the Room, When a Ventilating Fan Was Not Operated

Airborne particulates were collected at point A and point G in Fig. 1. Each period of time is as follows: 1, 11:07 (the previous day)–11:08; 2, 11:20–13:20 (broiling); 3, 13:29–15:29; 4, 15:35–17:35; 5, 17:41–19:41; 6, 19:52–11:00 (the following day). Particle size: □, ≥10 μm; ▨, 2.5 μm–10 μm; ■, ≤2.5 μm.

than the respective concentrations when ventilation was operated.

Figure 3 also shows the time course of the BaP concentration at a corner (point C) and the center (point B) of the room, without ventilation. The BaP concentration in the small particles during the cooking time (time-period 2) was lower at the center of the room than at the corner. BaP levels of these points, however, almost coincided after cooking (time-period 3). These results indicated that particles generated from mackerel pike moved to the center after progressing along the ceiling and the wall.

From these results, it was found that the PCI sampler could be used to measure PAH concentration in room-air. Except for cooking, there were no conceivable indoor-air-PAH sources such as kerosene heater use, incense stick burning, cigarette smoking, mosquito-repellent incense burning and so on. These sources contribute to indoor air pollution by BaP, to the extent of 1.6, 4.1, 8.2, 17.3 ng/m^3, respectively.[14] Considering that the mean concen-

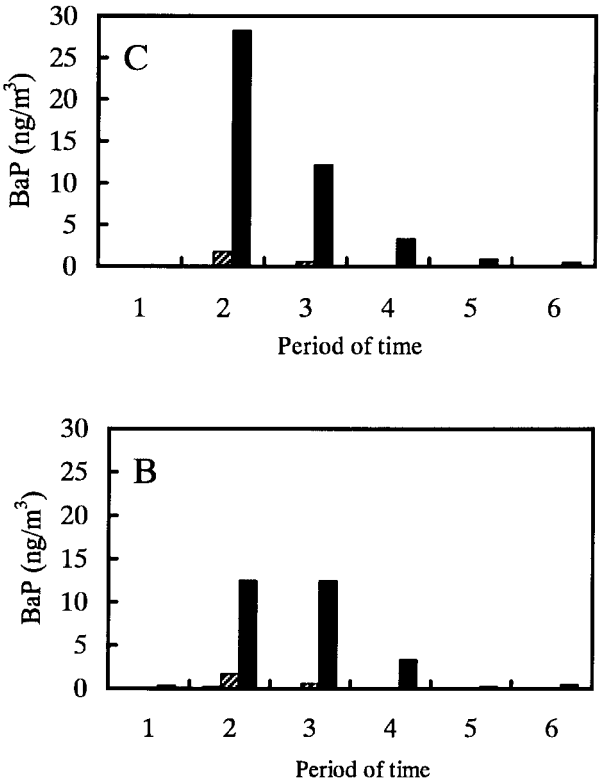

Fig. 3. BaP Concentration Changes in Size of Particles Over Time at the Center (B) and a Corner (C) of the Room, When a Ventilating Fan Was Not Operated

Airborne particulates were collected a t point B and point C in Fig. 1. Each time point, 1–6, was the same as in Fig. 2. Particle size: □, ≥10 μm; ▨, 2.5 μm–10 μm; ■, ≤2.5 μm.

tration of BaP in the indoor air of the Tokyo metropolitan area is 0.87 ng/m^3,[7] cooking is clearly a serious source of indoor air pollution by PAH. Moreover, the PAH level in indoor air was greatly different depending on operation/non-operation of a ventilation fan. In this model study, although the PAH levels in the large and medium particles were certainly increased as a result of cooking, the ratio of increase of the PAH level was overwhelmingly higher for small particles, by comparison. Therefore, it was confirmed that most of the PAHs generated were included in the small particles. Furthermore, the PAH concentration during cooking was higher at the corner opposite to the cooking stand than at the room center, despite the fact that the corner was farther away from the cooking stand. Therefore, the relation between monitoring points and room layout is very important, and it is fundamental to take layout into consideration when investigating indoor air contamination.

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