보건학석사 학위논문

Occupational Exposure to Particulate Matter, Polycyclic Aromatic Hydrocarbons and Heavy Metals among Workers at Korean BBQ Restaurant

육류직화구이 음식점 근로자의 입자상 물질, 중금속 및 다환방향족탄화수소 노출평가

2020년 8월

서울대학교 보건대학원

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이 논문을 보건학 석사 학위논문으로 제출함

2020년 6월

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조 혜 리의 보건학 석사 학위논문을 인준함

2020년 7월

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Abstract

Occupational Exposure to Particulate Matter, Polycyclic Aromatic Hydrocarbons and Heavy Metals among Workers at Korean BBQ Restaurant.

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Objective Korean BBQ restaurants use charcoal as a principal fuel source. During the process of meat roasting, cooking related emissions, such as: particulate matter (PM), heavy metals, and Polycyclic Aromatic Hydrocarbons (PAHs) are released, and are associated with adverse health effects. Although the emission characteristics of these substances have been previously studied in Korean BBQ restaurants, exposure assessments of workers to these substances remains under investigated. Thus, the purpose of this study was to evaluate the occupational exposure of restaurant workers to these emitted substances, as well as to determine workers' cancer risk potentials.

Methods Three Korean BBQ restaurants were investigated in this study. Personal and area samples were collected to determine concentrations of Total Suspended Particulates (TSP), heavy metals, and 16 PAHs in the halls, kitchens, and charcoal storages of each respective restaurant. Personal samples were taken from both hall and kitchen workers. For each restaurant, all samples were collected on different days for 8 hours per day, excluding break times. PAHs were analyzed using Gas chromatography mass spectrometry (GC-MS). Heavy metals were analyzed with inductively coupled plasma mass spectrometry (ICP-MS). Incremental Lifetime Cancer Risk (ILCR) was assessed in accordance with risk assessment guidance established by the US Environmental Protection Agency (US EPA).

Results The Geometric mean (GM) concentration of TSP by place was found in the order of: charcoal storage (196.30±1.97 $\mu g/m^3$), hall (86.61±1.76 $\mu g/m^3$), kitchen (44.39±2.60 $\mu g/m^3$), and outdoor air (15.37±2.47 $\mu g/m^3$). The TSP exposures of hall workers (141.45±2.08 $\mu g/m^3$) were about 3.8 times higher than those of kitchen workers (41.15±1.88 $\mu g/m^3$) ($p < 0.001$). The area and personal air concentrations of heavy metals were lower than standards set forth by Korea OEL. However, lead concentrations slightly exceeded Korean air quality standards of 0.5 ug/m^3. Cadmium and manganese exceeded WHO reference recommended standards of 0.005 ug/m^3 and 0.15 ug/m^3, respectively.

The average concentrations of total PAHs were not significantly different in the halls, kitchens, and charcoal storages. The average concentration of total PAHs of hall workers (1.77±1.89 μg/ m^3) was significantly higher than that of kitchen workers (0.23±2.04 μg/ m^3) ($p<0.001$). 2-ring and 3-ring PAHs were most commonly detected in the two groups. The ILCR average for PAHs was 9.5×10^{-5} for hall workers and 1.0×10^{-4} for kitchen workers. Among 25 workers, occupational exposure to PAH in two hall workers and one kitchen worker exceeded the reference value.

Conclusions Occupational exposures of TSP, PAHs, and heavy metals to workers in Korean grilled meat restaurant were measured. Hall workers igniting and carrying charcoal had higher TSP and PAHs exposure. The heavy metal exposure values of both hall and kitchen workers were below Korea's OEL. Hall workers were more likely to exceed the cancer risk reference value than kitchen workers. Therefore, it is necessary to manage these harmful substances in order to reduce worker exposure to the lowest possible level.

Keyword:	Cooking related emission, Charcoal, exposure assessment, Polycyclic aromatic hydrocarbons, particulate matters, heavy metals
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1. Introduction

More and more Koreans choose to eat out every year. According to a survey by Korea Agro-Fisheries & Food Trade Corporation (aT) in 2018, the average frequency of eating out for one person per month was 4.4 times for men and 2.5 times for women (aT.,2018). In addition, as of 2013, there were 28,475 restaurants nationwide, of which 5,623 were in Seoul (19.7%), and 4,241 restaurants were in Gyeonggi-do (14.9%). Collectively, they account for 38.3% of restaurants nationally (National Institute of Environmental Research, NIER, 2016). Accordingly, 220,813 workers (23.02%) and 224,798 workers (24.66%) were employed in Korean BBQ restaurants, respectively, in 2016 and 2017 (Food Information Statistics System, aT FIS, 2018).

These BBQ restaurants primarily use charcoal, briquettes, ignition coal (anthracite coal), gas, and electricity as ignition sources. Meat is often grilled either using a standard grill, or by using an iron plate placed on top of the ignition source. Due to the desired characteristics of grilled meat, the high-temperature heat source is located directly underneath the meat, so as to render the fat. When the oils from the fat come into contact with the fuel, incomplete combustion occurs and harmful substances such as volatile organic compounds (VOCs), aldehydes, carbon monoxide, fine dust, polycyclic aromatic hydrocarbons (PAHs), and heavy metals are generated (Park et al., 2015, Kim et al., 2017).

The fine dust generated by barbecued meat can be detrimental to human health, and have a tendency to linger in the air as suspended particulate matter. Some studies have shown that inhalation of fine dust can result inflammation of the airways, ultimately causing a reduction in lung function (Duan et al., 2013; Habre et al., 2014). Additionally, animal studies have implicated this fine dust in the development of cardiovascular disease and arteriosclerosis (Ballester et al., 2006; Dominici et al., 2006; Yatera et al., 2008).

A comparison of cooking methods for meat demonstrates the amount of fine dust generated during baking is approximately 878 $\mu g/m^3$, which is about 4 to 8 times higher than alternative methods such as boiling or frying (NIER, 2015). The amount of harmful substances generated by roasting pork belly was the highest for charcoal grilling as compared to using a gas or electric source (Habre et al., 2014). In charcoal bases grilling, PAH concentrations were signiificantly increased relative to other cooking methods as opposed to the concentrations of VOC and carbonyl compounds.

Korean BBQ restaurants generally use charcoal or anthracite coal as an ignition source. Previous studies have demonstrated that certain charcoal and anthracite coal products sold on the retail market contain heavy metal components such as lead, cadmium, nickel, chromium, and barium nitrate, and that these heavy metals are released into the air during their combustion processes (Kim et al., 2011). Additionally, the Ministry of Food and Drug Safety (MFDS) announced that nickel was detected at levels 4 times (0.4 mg / L) grater than the reference value (0.1 mg / L or less) in some commercial grill products (MFDS,2019).

Polycyclic aromatic hydrocarbons (PAHs) are compounds in which two or more benzene rings are combined. When introduced into the body, PAHs are metabolized into an unstable form of epoxide, and are known to be cytotoxic (Yang et al., 1977). Epidemiological studies have reported that PAHs are likely to cause lung cancer, skin cancer, leukemia, and stomach cancer. Consequently, the U.S. Environmental Protection Agency (EPA) regulates and manages 16 PAHs as priority pollutants (Avagyan et al., 2016; Abdel-Shafy and Mansour, 2016).

The process of roasting meats and fish at high temperatures, particularly using direct roasting approaches (e.g., charcoal fire), can result in human respiratory exposure to various harmful substances including fine dust, ultrafine dust, PAHs, and various heavy metals. One study concluded that in a comparative analysis of the concentration of fine dust and formaldehyde generated during the process of cooking *bulgogi*, the formaldehyde concentration increased from 0.01 ppm to 4 ppm (Lee et al., 2006).

Most previous studies related to the direct grilling of meat have only investigated the characteristics and concentration of air pollutants relative to the differences in the roasting method. However, there remains a significant lack of data regarding personal exposure assessments for workers in these establishments. Given the pre-existing data demonstrating correlations between meat grilling and air pollutants, it can be reasonably assumed that workers in Korean BBQ restaurants are constantly exposed to above normal levels of the above-mentioned hazardous materials. As such, it is imperative to investigate the occupational hazards associated with this profession.

Thus, the purpose of this study was to evaluate the occupational exposure of restaurant workers to major harmful substances, namely:

particulate matter, heavy metals, and PAHs. Furthermore, this study also aimed to correlate these exposures to corresponding cancer risks.

2. Materials and Methods

2.1. Study design

For the purposes of this study, three Korean BBQ restaurants were selected and analyzed. These restaurants were selected mainly because the cooking methods that they employed, mainly barbecuing, are known to generate substantial air pollutants. **Table 1** shows the characteristics and sample sizes in these three restaurants. In the case of Restaurants A and B, the charcoal is first ignited in the charcoal storage and then brought out into the hall. In the case of Restaurant C, the charcoal is ignited directly at the table in the hall, following which the lid is closed and the ventilation hood is positioned nearby.

All restaurants in this study followed the Special Act on the Improvement of Air Quality in the Seoul Metropolitan Area. The act aims to prevent air pollution caused by the emission sources of commercial BBQ restaurant by installing exhaust ventilation systems (hoods, ducts, and fans) above the stoves and ignition sources.

Personal samples were obtained from hall workers carrying food and charcoal in the halls, as well as igniting charcoal in the charcoal storage. Samples were also obtained from kitchen workers cooking in the kitchen.

Each participating worker wore two personal samplers: one for PAHs with TSP, and one for heavy metals. Area samples were collected to determine concentrations of Total Suspended Particulates (TSP), heavy metals, and PAHs in the hall, kitchen, and charcoal storage, where charcoal is stored and ignited (**Figure 1**).

Personal and area samples were collected at a height of 1.5 m to represent the breathing zone of a worker. Samples were collected for 8 hours, excluding break times (14:00-17:00) for 3 days. As a control, the outside air was measured of areas not affected by the ventilation facility, located within 10 m from the restaurant.

Table 1 Characteristic information of three Korean BBQ restaurant.

Restaurant /sampling condition	Restaurant A	Restaurant B	Restaurant C
Area (m^2)	191.01 m^2	184.06 m^2	184.06 m^2
Main cooking method	Barbecuing Pan-prying	Barbecuing Pan-prying	Barbecuing
Cooking fuel	Charcoal /LPG	Charcoal /LPG	Charcoal /LPG
Job type(n)			
Hall serving	2	2	2
Cooking	1	1	1
Ventilation system			
Local exhaust system			
	Hall(16) Kitchen(4)	Hall(12) Kitchen(6)	Hall(19) Kitchen(4)
Natural ventilation system			
	Door	Windows and door	Door
Sample size			
Personal sampling	9	9	7
Area sampling	12	12	12

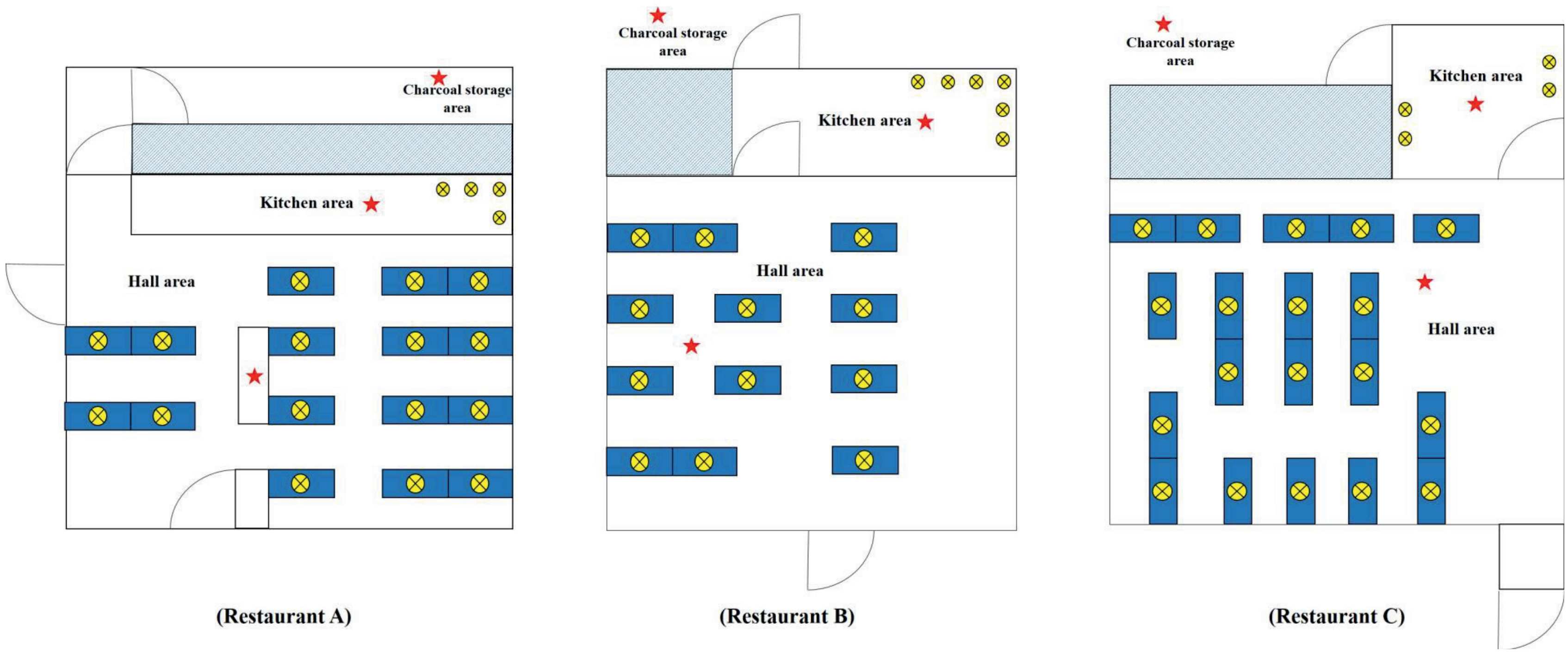

Figure 1. Restaurant schematic and sample location, the icons are the sampling locations (red star: hall, kitchen, charcoal storage area), cooking place using ignition source and ventilation system (the yellow circles), and the outside air is measured in a place that is not affected by restaurant ventilation system.

2.2. Sampling and analysis

2.2.1 Air sampling and analysis of TSP and PAHs

Polycyclic aromatic hydrocarbons (PAHs) 16 species are high priority pollutants specified by the EPA and include the following: Naphtalene, Acenaphthene, Phenanthrene, Anthracene, Fluoranthene, Pyrene, Benz(a)anthracene, Chrysene, Benzo(b)fluoranthene, Benzo(k)fluoranthene, Benzo(a)pyrene, Fluorene, Acenaphthylene, Debenz(a,h)anthracene, Benzo(ghi)perylene, and Indeno(1,2,3-C,D)pyrene(**Table A-1**). These species were measured and analyzed using the modified NIOSH 5506 method (NIOSH, 2003).

PTFE filter (37mm, Pore size 2 μm, PALL Corp, USA) in the three-piece 37mm closed face plastic cassette (Whatman Grade QM-A, 37mm ;Whatman, Maidstone, UK) and washed XAD-2 (100mg / 50mg, ORBO 43, Supelco) were connected in series. These were connected by air sampling pump (Gillian, U.S.A.) operating at an airflow rate of 2 ℓ / min. The samples were wrapped with foil during and after collection as not to expose them to sunlight (heat and ultraviolet light), and were then frozen.

To analyze the TSP, the PTFE filter was contained in a desiccator for more than 24 hours, and then weighed using an analytical balance with 0.01 μg sensitivity (Mettler XP6 Microbalance, Mettler Toledo, Hightstown NJ, USA). Before measuring the weight of the filter, static electricity was removed and the weight change according to the daily temperature and humidity change was corrected for using a blank filter. The weight was measured 3 times per sample, and the average value was used as the final weight concentration. The high flow rate pump was calibrated before and after sampling using a flow calibrator (Drycal, Defender 520-M, MesaLabs, USA).

After sampling, the PTFE filter and washed XAD-2 were transferred to an amber vial to prevent exposure to ultraviolet rays, and then 5 ml of acetonitrile in the case of a filter and 5 ml of toluene were injected by separating the front and back layers of washed XAD-2. Then, the lid was closed and sonicated for 30-60 minutes. The extracted sample was filtered using a PVDF 0.45 μm syringe filter. Quantitative analysis was performed on 16 PAHs using a gas chromatographer (Model 7890A, Agilent, USA) under the conditions specified by **Table 2**.

Table 2. The conditions of GC/MDS analyzed of PAHs

Parameter	Analytical conditions
Instrument	Gas Chromatography Mass Selective Detector (7890A/5975C, Agilent Technologies, USA)
Column	Agilent J&W DB-35ms (30 m×0.25 mm×0.25 ㎛, Aglient Tech., USA)
Injector Temperature	200 ℃
Injection Volume	1.0 ㎕
Split ratio	Splitless
Carrier gas	Helium (99.99% purity)
Detector Temperature	250 ℃
Oven condition	Initial 130℃ to 350℃ at 4℃/min (holding 4 min)

2.2.2 Air sampling and analysis of heavy metals

The sampling and analysis of heavy metals were based on the NIOSH method 7300 (NIOSH, 2016). MCE membrane filter paper (37 mm, Pore size 0.8 μm, Millipore, USA) was connected to the air sampling pump (Gillian, U.S.A.) operating at an airflow rate of 2 ℓ / min, and the same procedure, as described for PAH measurement, was performed. After sampling, the MCE filter was folded four times into a microwave vessel, and then 3 ml of 60% nitric acid solution (Sigma Aldrich, MO, USA) was added.

Samples were extracted using a membrane filter method stored in a library in a microwave (Model: MARS 6, CEM Corp., Matthews, NC, USA). The temperature was gradually raised to 200°C for 15 minutes and then maintained for 15 minutes. The pressure was set to 800 psi, and the power to 900–1050 W. After this process, the vessel was cooled in the microwave for 20 minutes and then placed at room temperature for 40 minutes. The extracted sample was diluted to 40 ml with 1% nitric acid solution, and quantitative analysis was performed using an inductively coupled plasma mass spectrometer (ICP-MS, Model: NexION 350D, Perkin Elmer Inc., Houston, TX, USA) under the conditions specified by **Table 3**.

Table 3. The conditions of ICP/MS analyzed of Heavy metals

Parameter	Analytical conditions
Nebulizer	Concentric glass nebulizer
Spray chamber	Glass cyclonic spray chamber
RF generator(W)	Power output : 500 W – 1,600 W
Argon flow rate	
Plasma gas (L/min)	18.00
Auxiliary gas (L/min)	1.20
Nebulizer gas (L/min)	0.96
Sampler cone (mm)	Nickel 1.0
Skimmer cone (mm)	Nickel 0.9
Hyper-Skimmer cone (mm)	Aluminum alloy 1.0
Vacuum	
Inter face(torr)	$< 2 \times 10^{-6}$
Quadrapole(torr)	$< 3 \times 10^{-8}$
Data acquisition	Peak hopping, 1 reading 20 sweep, 3 replicate
Measurement mode	Quantification mode

2.3 Quality assurance and Quality controls(QA/QC)

Quality assurance and Quality controls (QA/QC) for PAHs and heavy metals was performed.

The limit of detection (LOD) of 16 PAHs and heavy metals were calculated in different ways. The LOD of PAHs, as well as the internal standard method, were applied to the quantification of the substance to be analyzed, and a calibration curve using the multi-standard mixture (AccuStandard®) was prepared using 6 standard samples (0.05 to 10 μg/L). The correlation coefficient (r^2) was more than 0.99 linearity, and multiplied by 3.3 (FDA.,1997). The results are described in (**Table A-2**).

The LODs of heavy metals was determined by threefold standard deviations from seven replicates at the lowest level of the standard solution (1 to 5 μg/L), and yielded a correlation coefficient (r^2) of greater than 0.99 and showed linearity (EPA, 2015). The values are: Mn 0.061, Ni 0.012, Cr 0.096, Fe 0.010, Pb 0.067, Cd 0.114, As 0.011 μg/sample.

2.4. Estimation of Cancer Risks

In this study, cancer risks were estimated by considering the inhalation exposures of workers in the three restaurants. For each worker, Incremental Lifetime Cancer Risk (ILCR) was assessed in accordance with risk assessment guidelines established by the US Environmental Protection Agency (US EPA, 2011; 1989). The ILCR was calculated using the equation shown blow:

$$\text{ILCR} = \text{Intake} \cdot \text{SF} \quad (1)$$

$$\text{Intake} = (\text{C} \cdot \text{IR} \cdot \text{ET} \cdot \text{EF} \cdot \text{ED}) / (\text{BW} \cdot \text{LT}) \quad (2)$$

where Intake is the lifetime average daily dose(μg/kg-day), C is the concentration (μg / m^3) of a pollutant in the air from each restaurants, IR is the inhalation rate (0.675 m^3/ hour for males and 0.543 m^3/hour for females) (NIER, 2019), ET is exposure time (hours/day), EF is exposure frequency(days/year), ED is exposure duration(years), BW is body weight(kg), LT is averaging time (period over which exposure is averaged-day), and SF is slope factor ([μg/kg-day]$^{-1}$).

The slope factors for PAHs, nickel, lead, cadmium, arsenic were obtained from 0.0039, 0.00091, 0.00042, 0.015 and 0.012, respectively (US EPA, 2016). According to the US EPA, an ILCR lower than 1×10^{-6}

can be regarded as negligible, and an ILCR above 1×10^{-4} is likely to be harmful to human beings. The ILCR value with in a range of 1×10^{-6} to 1×10^{-4} indicates a tolerable risk for the public (US EPA, 2001).

2.5. Statistical analysis

Data from samples of TSP, PAHs, and heavy metals were divided according to the personal and stationary measurements summarized for each sampling location and restaurant. The air sample concentrations are expressed in terms of the arithmetic mean (AM), standard deviation (SD), geometric mean (GM), geometric standard deviation (GSD), and range (min-max). The results of the Shapiro-Wilk test confirmed that the TSP, PAH, heavy metal, and ILCR data were log-normally distributed. Comparison of measured air concentrations of TSP, PAHs and heavy metals between the three restaurants was performed using ANOVA, and the Bonferroni method was used for post-hoc analysis. Welch two sample t-test was performed to compare personal concentrations of TSP, heavy metals, and PAHs between hall and kitchen workers. Comparison of the ILCR between the hall and kitchen workers was made by a Mann-Whitney U test. $p < 0.05$ was considered statistically significant. All statistical analysis was performed using R software v.3.5 (R Development Core Team, Vienna, Austria).

3. Results

3.1. Personal and area concentrations

3.1.1 Total Suspended Particulates (TSP)

Table 4 lists the TSP concentrations of personal and area samples measured in the three Korean BBQ restaurants. The geometric mean (GM) concentrations of personal and area samples were 92.7±2.8 μg/m^3 (AM±SD: 147.6±147.3 μg/m^3) and 89.3±2.6 μg/m^3 (AM±SD: 136.7±172.9 μg/m^3), respectively. The T-test results confirmed that the GM concentrations of personal and area samples were not statistically different ($p>0.05$). This shows that area exposure can be used as a surrogate of personal exposure.

The difference in the GM concentration of the workers in the three restaurants was not statistically significant ($p>0.05$). At the three restaurants A, B, and C, the TSP concentration of hall workers was 3.0 times, 5.5 times, and 2.1 times higher than those of kitchen workers, respectively. This confirmed that there was a statistical difference ($p<0.001$). In addition, as shown in **Figure 2-(a)**, the GM concentration of TSP of hall workers was 141.4±2.1 μg/m^3 (AM±SD: 181.3±139.1 μg/m^3), which was 3.4 times higher than that of kitchen workers 41.2 ±1.9 μg/m^3 (AM±SD: 47.9±24.2 μg/m^3). These values were significantly different ($p<0.001$). The hall workers igniting and carrying out charcoal were exposed to a maximum of 578.7 μg/m^3, which was a greater exposure relative to kitchen workers.

The GM concentrations of TSP excluding the outside air value of the three restaurants A, B and C were 127.8 ±1.55 μg/m^3,72.71 ± 3.99 μg/m^3 and 81.22±2.22 μg/m^3 (AM±SD: 140.6±74.73 μg/m^3, 171.38±289.41 μg/m^3, 104.41±70.02 μg/m^3), respectively. The difference in the means of the three restaurants was not statistically significant ($p>0.05$). However, among the three groups, the lowest hall concentration was found in Restaurant C, and the lowest kitchen concentration was found in Restaurant B. In both of these cases, natural and local ventilation were performed simultaneously **(Table 4)**.

In **Figure 2-(b)**, the GM concentrations of TSP showed high average concentrations in the order of charcoal storage, hall, and kitchen, measuring in at 86.6 ±1.8 μg/m^3, 44.4 ± 2.6 μg/m^3, and 196.3 ± 2.0 μg/m^3 (AM±SD:97.7±43.3 μg/m^3, 62.0±45.2 μg/m^3, 256.7±262.1 μg/m^3), respectively. Compared to the outside air, it was 5.6 times, 2.9 times, and 12.8 times higher, respectively.

Table 4. TSP concentration in air (µg/㎥) determined via area and personal sampling in three Korean BBQ restaurants

Restaurant	Location	N	AM±SD(µg/m^3)	GM(GSD)	Range(µg/m^3)	p-value
Personal						
A	Hall	6	208.6±101.6	190.0(1.6)	122.9-342.9	p >0.05[a]
	Kitchen	3	63.8±9.7	63.3(1.2)	56.0-74.6	
B	Hall	6	208.7±195.8	145.9(2.6)	48.5-578.7	
	Kitchen	3	32.8±25.0	26.7(2.2)	12.4-60.7	
C	Hall	4	99.2±60.9	86.8(1.8)	46.3-184.9	
	Kitchen	3	47.0±30.3	41.2(1.9)	24.4-81.4	
Total		25	147.6±147.3	92.7(2.8)	12.4-578.7	

Area						
A	Hall	3	118.9±37.1	115.3(1.4)	91.6-161.2	
	Kitchen	3	92.7±14.6	91.9(1.2)	78.0-107.1	
	Storage	3	210.3±96.5	197.1(1.6)	140.6-320.4	
	Outdoor	3	41.8±11.3	40.8(1.3)	31.2-53.8	
B	Hall	3	105.6±23.5	103.8(1.3)	79.9-126.0	
	Kitchen	3	17.8±11.6	15.6(1.8)	10.5-31.2	
	Storage	3	390.7±469.4	237.2(3.3)	114.7-932.7	$p > 0.05$ [b]
	Outdoor	3	10.1±4.0	9.6(1.5)	6.5-14.4	
C	Hall	3	68.5±59.45	54.3(2.2)	33.2-137.1	
	Kitchen	3	75.6±56.6	61.0(2.3)	26.0-137.3	
	Storage	3	169.1±58.4	161.8(1.5)	107.4-223.5	
	Outdoor	3	12.8±13.0	9.3(2.6)	5.2-27.8	
Total[c]		27	136.7±172.9	89.3(2.6)	10.5-932.7	

[a] The GM of TSP concentration difference among workers working in each restaurant.
[b] The GM of TSP concentration difference among each restaurants.
[c] The outdoor value was excluded.

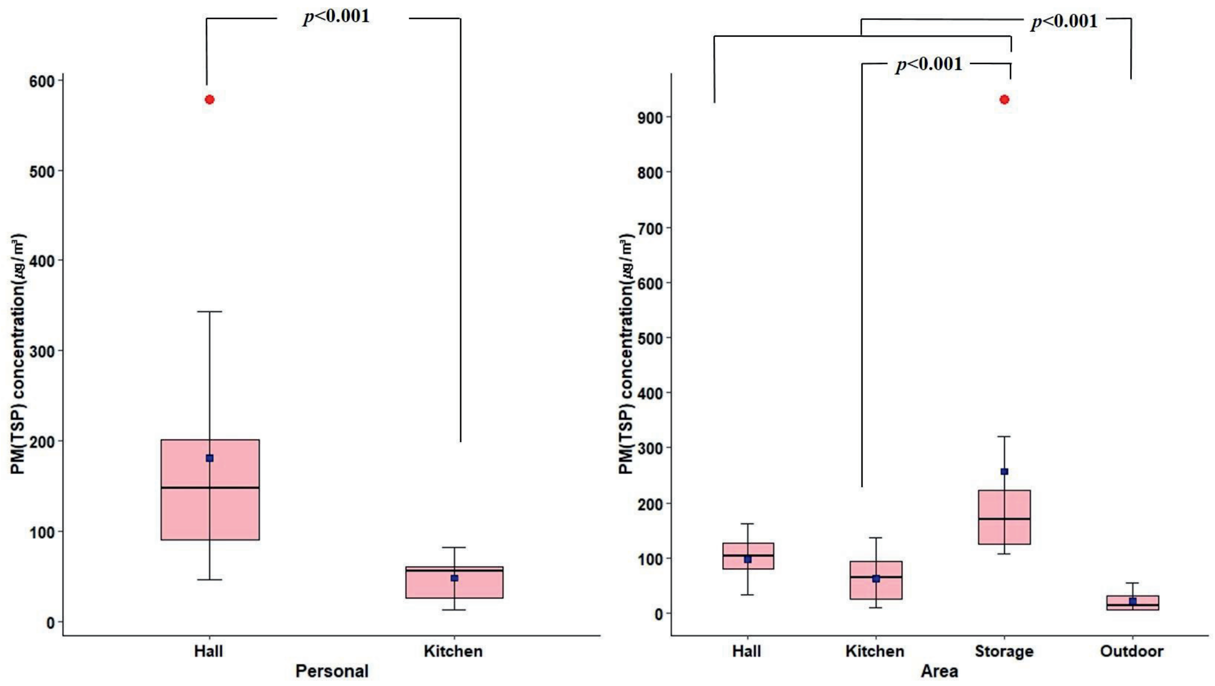

Figure 2 Box plots of TSP concentration in air (μg/㎥) determined via area and personal by (a) hall, kitchen, storage and Outdoor and (b) hall workers and kitchen workers (p <0.001). The TSP concentration is geometric mean. Values shown are median (line within box), mean (closed blue square within box), 25th and 75th percentiles (bottom and top of box, respectively), minimum (lower bars on whisker), maximum (upper bars on whisker), and outliers (red dots).

3.1.2 Heavy metals

Figure 3 and **Figure 4** show the personal and area exposure concentration of heavy metals by location. The concentration of heavy metals in hall and kitchen workers was below Korea’s occupational exposure limits (OEL). In particular, nickel, chromium, barium, lead, and cadmium, which are known to be emitted into the air during charcoal burning, did not exceed OEL. However, levels of lead were higher than Korea's Air quality standards (0.5 μg/m^3) by 2.1 times and 2.3 times in the hall workers (1.07±1.18 μg/m^3) and the kitchen workers (1.13±1.01 μg/m^3), respectively. Those of cadmium and manganese exceeded the World Health Organization (WHO) reference recommendation standard (0.005 μg/m^3 for cadmium and 0.15 μg/m^3 for manganese). In the case of cadmium, hall worker (0.04±0.02 μg/m^3) and kitchen worker (0.03±0.01 μg/m^3) exposures were 7.3 times and 5.6 times higher, respectively. In the case of manganese, hall worker (13.38±27.79 μg/m^3) and kitchen worker (1.19±0.82 μg/m^3) exposures were 89.2 times and 7.9 times higher than the standard, respectively. **(Table A-4 & Table A-5)**.

Table 5 shows the concentration of personal heavy metals for each of the three Korean BBQ restaurants. For manganese, cadmium, barium, and chromium, statistical differences were confirmed for each restaurant ($p<0.005$ and $p<0.05$). In the case of nickel, workers at the three restaurants had similar exposure levels. It was confirmed that nickel had the lowest exposure value in Restaurant C ($p<0.05$), and other heavy metals did not show statistical differences by each restaurant (**Table 6**).

The concentration of heavy metals in area samples by location was also less than that of OEL **(Figure 4)**. However, it was confirmed that there is a statistically significant difference from the personal exposure concentration ($p<0.05$), although there are differences by location and element. In particular, in the case of manganese, it was mainly detected by the charcoal storage and in hall workers. As such, it can be inferred that the hall workers were exposed in the process of igniting or carrying the charcoal. In addition, nickel and cadmium were mainly detected in the hall, and barium and lead were detected in the hall and the charcoal storage. Through this, it was found that the restaurants in this study use products containing these elements. However, in the case of arsenic, there were no significant differences in the values of personal samples and area samples ($p>0.05$).

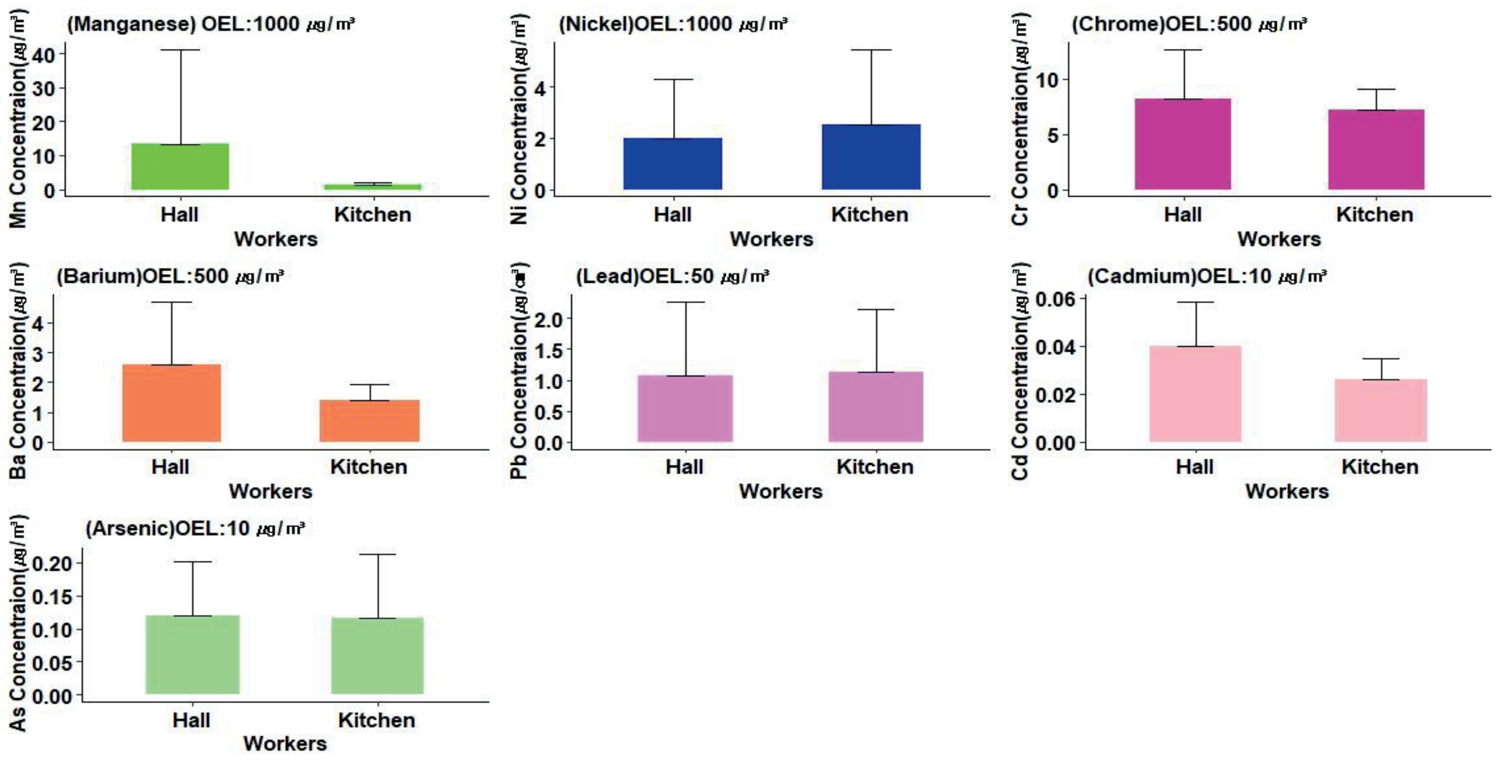

Figure 3 Bar plots of Heavy metals concentration in air (μg/㎥) determined via personal sample in three Korean BBQ restaurants. Error bars show the standard deviation. Korea's Air quality standards for lead: 0.5 μg/m^3, WHO reference recommendation standard for cadmium and manganese: 0.005 μg/m^3, 0.15 μg/m^3, respectively.

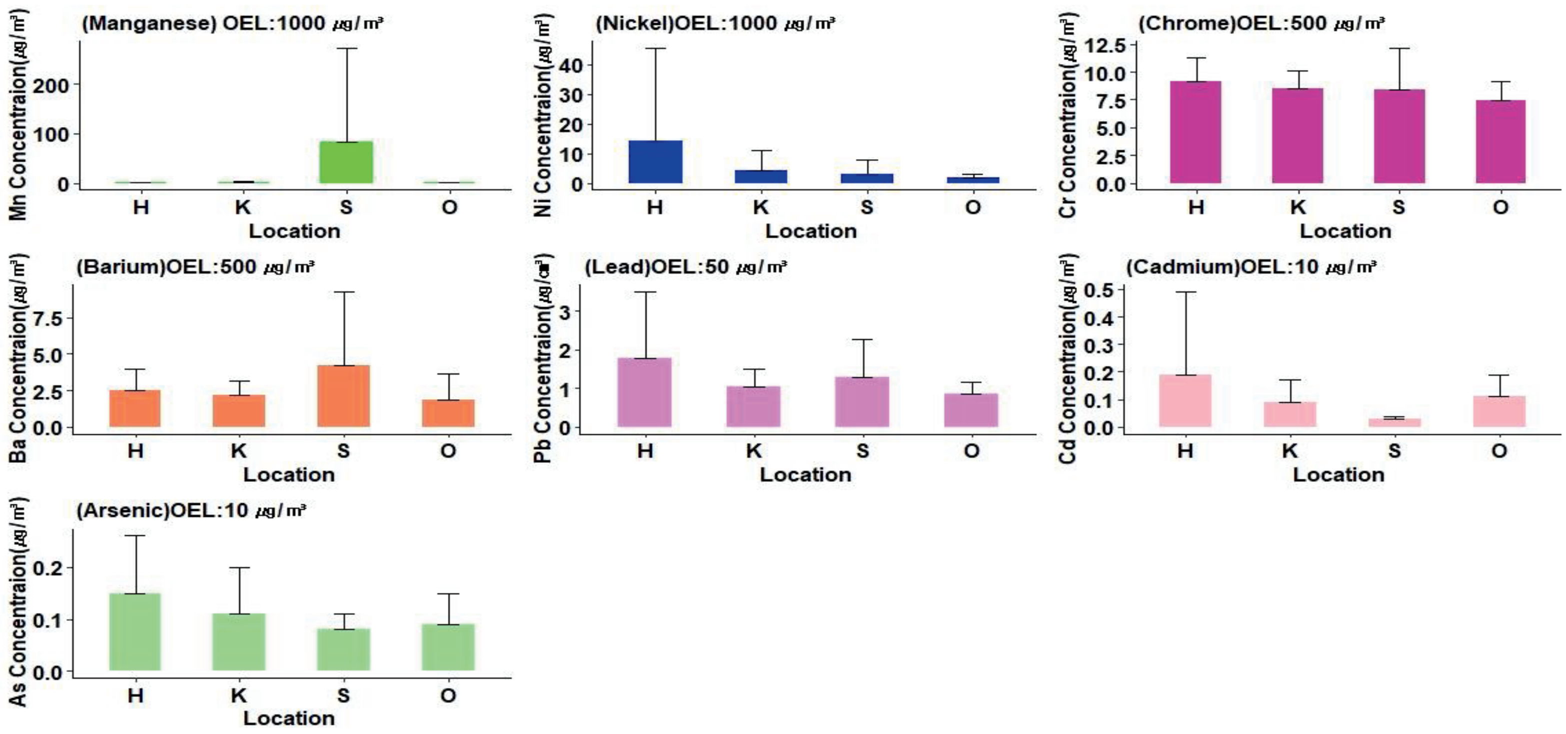

Figure 4 Bar plots of Heavy metals concentration in air (μg/㎥) determined via area sample in three Korean BBQ restaurants. Error bars show the standard deviation. Korea's Air quality standards for lead: 0.5 μg/m^3, WHO reference recommendation standard for cadmium and manganese: 0.005 μg/m^3, 0.15 μg/m^3, respectively.

Table 5 Heavy metals concentration in air (μg/㎥) determined via personal sampling in three Korean BBQ restaurants

Restaurant	Location		Species/Chemical(μg/㎥)						
			Mn**	Ni	Cr*	Ba*	Pb	Cd**	As
	Personal								
A	**Hall(N=6)**	Mean±SD	1.56±0.98	1.39±0.73	8.34±7.41	2.01±1.47	1.46±1.89	0.04±0.02	0.14±0.07
		GM(GSD)	1.24(2.22)	2.21(1.82)	6.76(1.87)	1.62(2.06)	0.90(2.71)	0.03(2.24)	0.12(1.92)
		Range	0.39-2.45	0.49-2.42	4.40-23.37	0.57-4.59	0.27-5.27	0.01-0.07	0.04-0.24
	Kitchen(N=3)	Mean±SD	1.06±0.87	1.40±0.53	5.09±0.43	1.85±0.54	0.90±0.18	0.03±0.01	0.13±0.10
		GM(GSD)	0.85(2.23)	1.33(1.44)	5.07(1.09)	1.80(1.33)	0.89(1.22)	0.03(1.57)	0.11(2.36)
		Range	0.42-2.05	0.97-1.99	4.66-5.51	1.40-2.45	0.74-1.10	0.02-0.04	0.04-0.24
B	**Hall(N=6)**	Mean±SD	25.21±36.92	1.54±0.51	7.86±2.40	4.04±2.51	0.94±0.32	0.01±0.00	0.13±0.10
		GM(GSD)	8.85(7.80)	1.47(1.41)	7.49(1.45)	3.52(1.83)	0.89(1.36)	0.01(1.30)	0.11(1.27)
		Range	0.41-98.05	0.95-2.46	3.98-10.10	1.71-8.74	0.48-1.47	0.01-0.02	0.07-0.33
	Kitchen(N=3)	Mean±SD	1.39±1.01	2.12±0.88	8.21±1.48	1.42±0.42	0.91±0.13	0.01±0.01	0.07±0.02
		GM(GSD)	0.67(2.24)	1.67(1.65)	8.55(1.19)	1.19(1.35)	0.87(1.17)	0.01(1.89)	0.07(1.32)
		Range	<LOD-2.10	1.11-2.72	7.32-9.91	1.02-1.86	0.76-1.00	0.01-0.02	0.05-0.09

C	**Hall(N=4)**	Mean±SD	<LOD	3.62±4.44	8.34±1.28	1.31±0.74	0.69±0.59	0.01±0.01	0.07±0.04
		GM(GSD)	<LOD	2.09(3.35)	8.27(1.16)	1.15(1.85)	0.51(2.45)	0.01(3.10)	0.06(1.95)
		Range	<LOD	0.54-10.22	7.39-10.22	0.66-1.98	0.24-1.48	0.00-0.03	0.02-0.13
	Kitchen(N=3)	Mean±SD	<LOD	4.06±5.23	8.28±1.58	0.95±0.09	1.57±1.89	0.01±0.01	0.14±0.15
		GM(GSD)	<LOD	2.09(4.18)	8.18(1.21)	0.94(1.10)	0.88(3.88)	0.01(2.60)	0.09(2.96)
		Range	<LOD	0.61-10.08	6.77-9.92	0.88-1.05	0.25-3.73	<LOD-0.02	0.04-0.31
Total(N=25)		Mean±SD	9.80±23.75	2.19±2.47	7.81±3.79	2.17±1.77	1.09±1.10	0.02±0.02	0.12±0.08
		GM(GSD)	2.31(4.79)	1.59(2.07)	7.23(1.46)	1.72(1.95)	0.82(2.85)	0.01(2.51)	0.09(1.97)
		Range	<LOD-98.05	0.49-10.22	3.98-23.37	0.57-8.74	0.24-5.27	<LOD-0.07	0.02-0.33

* The GM of heayv metals concentration difference among workers working in each restaurant ($p<0.005$).
** The GM of heayv metals concentration difference among workers working in each restaurant ($p<0.05$).
Abbreviation : Mn= Manganese, Ni=Nickel, Cr=Chromium, Ba=Barium, Pb= Lead, Cd = Cadmium, As= Arsenic.

Note : (1) Korea's occupational exposure limits : Mn=1000 μg/㎥, Ni=1000 μg/㎥, Cr=500 μg/㎥, Ba=500 μg/㎥, Pb= 50 μg/㎥, Cd = 10 μg/㎥, As= 10 μg/㎥.
(2) Korea's Air quality standards = 0.5 μg/m^3, WHO recommendation standard = 0.005 μg/m^3 for cadmium and 0.15 μg/m^3 for manganese.

Table 6 Heavy metals concentration in air (μg/㎥) determined via area sampling in three Korean BBQ restaurants

Restaurant	Location		Species/Chemical(μg/㎥)						
			Mn	Ni*	Cr	Ba	Pb	Cd	As
	Area								
A	Hall(N=3)	Mean±SD	1.23±1.91	35.53±54.08	7.58±2.81	3.65±1.47	3.39±2.35	0.35±0.38	0.24±0.09
		GM(GSD)	1.04(2.01)	11.85(6.39)	7.53(1.42)	3.43(1.57)	2.59(2.77)	0.19(4.67)	0.23(1.42)
		Range	0.60-2.28	3.05-97.96	5.48-10.95	2.09-4.99	0.81-5.40	0.04-0.77	0.17-0.34
	Kitchen (N=3)	Mean±SD	1.64±1.18	1.21±0.69	7.52±1.18	2.42±1.59	1.05±0.56	0.07±0.09	0.15±0.11
		GM(GSD)	1.36(2.15)	1.06(1.96)	7.46(1.17)	1.86(2.74)	0.92(1.99)	0.04(4.01)	0.11(3.14)
		Range	0.64-2.94	0.50-1.87	6.47-8.81	0.58-3.43	0.42-1.49	0.01-0.18	0.03-0.24
	Storage (N=3)	Mean±SD	0.82±0.74	1.50±0.41	5.01±1.06	1.47±0.14	0.59±0.11	0.02±0.00	0.09±0.04
		GM(GSD)	0.63(2.93)	1.47(1.32)	4.95(1.24)	1.47(1.10)	0.59(1.21)	0.02(1.02)	0.09(1.51)
		Range	0.29-1.34	1.21-1.78	4.25-5.76	1.37-1.57	0.51-0.67	0.02-0.02	0.07-0.12
	Outdoor (N=3)	Mean±SD	0.83±0.21	2.29±2.06	6.76±2.76	3.34±2.81	0.80±0.45	0.07±0.08	0.14±0.08
		GM(GSD)	0.81(1.28)	1.61(3.04)	6.42(1.47)	2.51(2.66)	0.69(2.06)	0.04(4.14)	0.13(1.72)
		Range	0.65-1.06	0.50-4.54	4.66-9.89	0.91-6.41	0.30-1.19	0.01-0.16	0.08-0.23

B	**Hall(N=3)**	Mean±SD	3.06±1.63	9.77±1.46	1.73±2.19	1.11±1.16	0.04±0.49	0.14±0.00	0.06±0.12
		GM(GSD)	1.07(3.76)	2.81(1.69)	9.62(1.24)	1.51(1.85)	1.05(1.50)	0.04(0.00)	0.11(2.26)
		Range	0.42-2.72	1.61-4.53	8.43-12.30	1.01-3.06	0.80-1.67	LOD-0.04	0.06-0.28
	Kitchen (N=3)	Mean±SD	8.27±0.00	8.44±11.17	2.15±1.28	1.21±0.60	0.01±0.39	0.07±0.00	0.82±0.02
		GM(GSD)	2.28(0.00)	4.09(1.20)	8.37(1.17)	2.09(1.33)	1.16(1.44)	0.01(0.00)	0.07(1.33)
		Range	<LOD-2.28	1.52-21.16	7.15-9.70	1.55-2.75	0.76-1.46	<LOD-0.01	0.05-0.09
	Storage (N=3)	Mean±SD	1.66±325.58	10.43±0.48	6.54±4.99	1.18±8.11	0.03±0.30	0.10±0.00	4.22±0.04
		GM(GSD)	68.36(15.30)	1.61(1.38)	9.67(1.61)	3.63(3.81)	1.15(1.27)	0.03(0.00)	0.09(1.49)
		Range	9.94-470.38	1.11-1.99	6.22-15.95	1.17-15.87	0.98-1.52	<LOD-0.03	0.06-0.13
	Outdoor (N=3)	Mean±SD	1.73±0.00	1.34±0.26	7.52±1.09	1.18±0.37	0.78±0.17	<LOD	0.06±0.02
		GM(GSD)	1.73(0.00)	1.33(1.20)	7.47(1.16)	1.13(1.40)	0.77(1.24)	<LOD	0.06(1.36)
		Range	<LOD-1.73	1.15-1.64	6.36-8.53	0.18-1.51	0.64-0.97	<LOD	0.05-0.09
C	**Hall(N=3)**	Mean±SD	1.03±0.00	3.60±1.45	9.81±1.68	2.15±1.45	0.86±0.40	0.02±0.01	0.07±0.05
		GM(GSD)	1.03(0.00)	3.42(1.45)	9.71(1.19)	1.77(2.29)	0.78(1.79)	0.02(1.36)	0.06(2.09)
		Range	<LOD-1.03	2.70-5.27	8.12-11.47	0.71-3.60	0.40-1.14	0.02-0.02	0.03-0.12
	Kitchen (N=3)	Mean±SD	<LOD	2.73±1.72	9.99±2.29	1.88±0.59	0.82±0.57	0.02±0.02	0.12±0.13
		GM(GSD)	<LOD	2.45(1.97)	9.86(1.26)	1.83(1.38)	0.72(2.14)	0.02(3.31)	0.08(4.03)
		Range	<LOD	1.52-3.95	8.38-11.61	1.46-2.29	0.42-1.23	0.01-0.04	0.03-0.22

Storage (N=3)	Mean±SD	10.64±2.25	5.57±8.01	8.56±2.64	3.72±2.72	1.85±1.50	0.03±0.01	0.07±0.02
	GM(GSD)	10.52(1.24)	2.31(5.11)	8.27(1.38)	2.71(3.04)	1.10(4.59)	0.02(1.71)	0.06(1.40)
	Range	9.05-12.23	0.70-14.81	5.84-11.12	0.76-6.11	0.19-3.11	0.02-0.04	0.04-0.08
Outdoor (N=3)	Mean±SD	<LOD	1.90±0.72	7.90±1.36	1.00±0.28	0.96±0.42	<LOD	0.07±0.02
	GM(GSD)	<LOD	1.79(1.56)	7.82(1.20)	0.97(1.37)	0.91(1.49)	<LOD	0.07(1.29)
	Range	<LOD	1.07-2.39	6.33-8.69	0.68-1.17	0.72-1.44	<LOD	0.05-0.09
Total(N=27)[a]	Mean±SD	26.13±104.6	5.93±16.76	8.34±2.43	2.66±2.79	1.25±1.06	0.09±0.17	0.11±0.08
	GM(GSD)	2.03(5.17)	2.35(2.82)	8.02(1.33)	1.94(2.12)	0.98(1.99)	0.04(3.34)	0.09(1.92)
	Range	<LOD-470.38	<LOD-97.96	<LOD-15.95	<LOD-15.87	<LOD-5.40	<LOD-0.77	<LOD-0.34

[a] The outdoor value was excluded.

[*] The GM of heayv metals concentration difference among each restaurant ($p<0.05$).

Abbreviation : Mn= Manganese, Ni=Nickel, Cr=Chromium, Ba=Barium, Pb= Lead, Cd = Cadmium, As= Arsenic.

Note : (1) Korea's occupational exposure limits : Mn=1000 μg/㎥, Ni=1000 μg/㎥, Cr=500 μg/㎥, Ba=500 μg/㎥, Pb= 50 μg/㎥, Cd = 10 μg/㎥, As= 10 μg/㎥. (2) Korea's Air quality standards = 0.5 μg/m^3, WHO recommendation standard = 0.005 μg/m^3 for cadmium and 0.15 μg/m^3 for manganese.

3.1.3 Polycyclic aromatic hydrocarbons (PAHs)

Figure 5 shows the personal average concentrations of total PAHs for hall workers (n=16) and kitchen workers (n=9). The GM concentration of total PAHs for hall workers and kitchen workers was 1.77±1.89 μg/ m^3 and 0.23±2.04 μg/ m^3, respectively ($p<0.001$), and most of the 2-ring and 3-ring PAHs were detected in these two groups. This shows that hall workers who directly handle charcoal are at a greater risk for exposure to PAHs than kitchen workers. The personal PAH concentration was 0.27±3.46 μg/ m^3, and the area PAH concentration excluding the outdoor air concentration was 0.25±4.33 μg/ m^3, which confirmed that there was no statistical difference ($p>0.05$).

The GM concentrations of total PAHs were not significantly different in hall, kitchen, and charcoal storage (0.21± 4.81μg/ m^3, 0.30 ±2.69μg/ m^3 and 0.27 ±4.66μg/ m^3, respectively), and showed a statistically significant difference compared to the concentration of outside air (0.10±2.27 μg/ m^3) ($p<0.001$). Compared to other PAHs, most of the 2-ring and 3-ring PAHs, which have lower toxic equivalent factors, were detected. The most potent carcinogen, benzo(a)pyrene was detected in the halls and charcoal storages (**Table 5**).

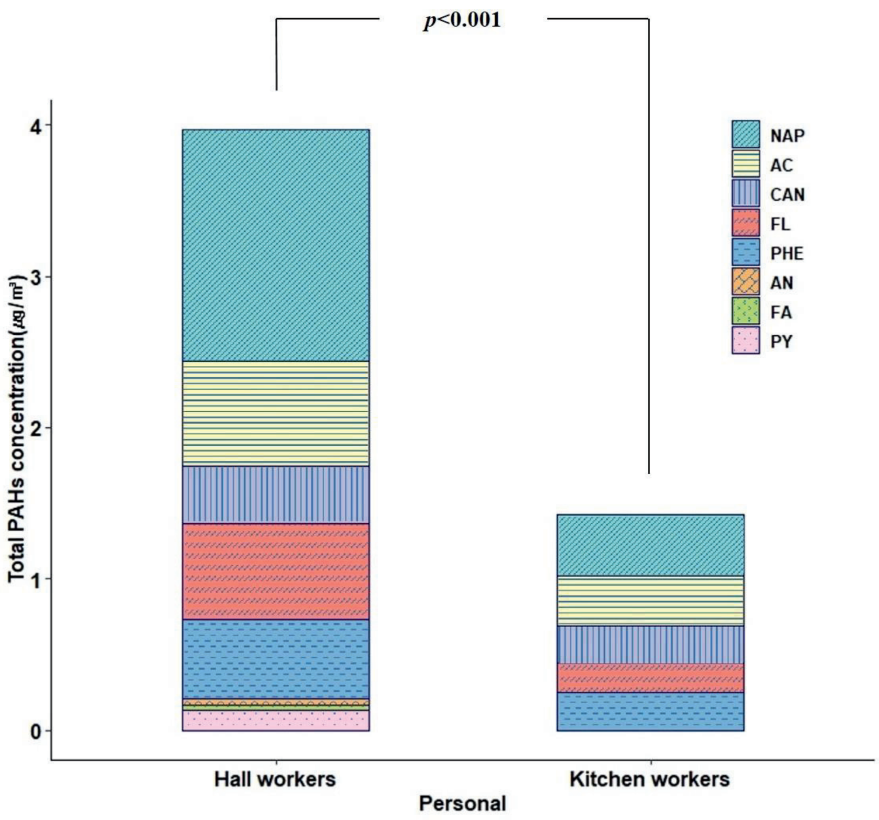

Figure 5. PAHs concentration in air (μg/㎥) determined via personal sampling by location in three Korean BBQ restaurants.

Table 7. PAHs concentration in air (㎍/㎥) determined via area sampling by location in three Korean BBQ restaurants.

Species /Chemical	Hall(N=9)			Kitchen(N=9)			Storage(N=9)		
	Mean ±SD	GM (GSD)	Range	Mean ±SD	GM (GSD)	Range	Mean ±SD	GM (GSD)	Range
NA	2.27±1.29	1.90(1.96)	0.71-4.49	0.85±0.33	0.78(1.54)	0.32-1.48	2.22±0.98	2.04(1.52)	1.10-4.23
ACY	1.19±0.75	0.96(2.10)	0.22-2.58	0.46±0.27	0.35(2.59)	0.04-0.88	1.25±0.82	1.03(1.95)	0.35-2.89
ACE	0.59±0.24	0.54(1.65)	<LOD -0.90	0.28±0.18	0.23(2.08)	<LOD-0.56	0.69±0.52	0.52(2.40)	0.13-1.88
FLU	0.76±0.44	0.66(1.77)	<LOD -1.67	0.44±0.13	0.42(1.35)	<LOD-0.62	0.85±0.71	0.65(2.25)	0.13-2.61
PHE	0.34±0.28	0.25(2.35)	0.08-0.82	0.40±0.30	0.31(2.38)	<LOD-0.96	0.79±0.32	0.73(1.61)	0.24-1.41
AN	0.42±0.24	0.34(2.31)	<LOD -0.73	0.13±0.15	0.07(3.45)	<LOD-0.30	0.48±0.26	0.42(1.74)	0.21-0.92
FA	0.40±0.36	0.23(3.52)	<LOD -1.02	0.31±0.24	0.22(2.85)	<LOD-0.66	0.54±0.24	0.46(2.12)	0.07-0.93
PY	0.39±0.18	0.33(2.06)	<LOD -0.64	0.33±0.39	0.16(3.90)	<LOD-1.00	0.86±0.16	0.85(1.20)	0.67-1.20

BaA	0.10±0.15	0.05(3.02)	<LOD -0.48	<LOD			0.43±0.16	0.40(1.50)	0.21-0.70
CHR	0.10±0.13	0.07(2.38)	<LOD -0.43	<LOD			0.40±0.23	0.34(1.98)	0.08-0.88
BbF	0.03±0.01	0.03(1.37)	<LOD -0.04	<LOD			0.10±0.06	0.08(2.12)	0.03-0.19
BkF	0.03±0.00	0.03(1.10)	<LOD-0.03	<LOD			0.46±0.36	0.28(3.34)	0.04-0.90
BaP	0.03±0.00	0.03(1.09)	<LOD -0.04	<LOD			0.18±0.25	0.08(4.52)	<LOD-0.84
IP	0.02±0.01	0.02(1.31)	<LOD -0.02	<LOD			0.03±0.01	0.03(1.37)	<LOD-0.03
DahA	0.02±0.01	0.02(1.76)	<LOD-0.04	<LOD			0.02±0.00	0.02(1.24)	<LOD-0.03
BghiP	0.03±0.00	0.03(1.06)	<LOD-0.03	<LOD			0.03±0.01	0.02(2.11)	<LOD-0.04
Total PAH	0.56±0.79	0.21(4.81)	<LOD-4.49	0.44±0.32	0.30(2.69)	<LOD-1.48	0.60±0.68	0.27(4.66)	<LOD-4.23

Abbreviation: 16 priority control PAHs : NAP= Naphthalene, ACY=Acenaphthylen, ACE=Acenaphthene, FLU=Fluorene, PHE=Phenanthrene, ANT=Anthracene, FLUA=Fluoranthere, PYR=Pyrene, BaA=Benzo(a)anthracene, CHR=Chrysene,BaF=Benzo(b)fluoranthene, BkF=Benzo(k)fluoranthene, BaP=Benzo(a)pyrene, IND=Ideno(1,2,3)pyrene, DBahA= Dibenz(a,h)anthracene, BghiP=Benxo(g,h,i)perylene

3.2. Incremental lifetime cancer risk (ILCR)

The ILCR for nickel, lead, cadmium and arsenic ranged from 4.9×10^{-7} to 3.2×10^{-5}, 9.9×10^{-8} to 5.9×10^{-6}, 4.1×10^{-7} to 7.0×10^{-6} and 2.9×10^{-7} to 3.5×10^{-5}, respectively. The ILCR for total PAHs ranged from 4.9×10^{-6} to 4.9×10^{-4}. In the case of heavy metals, no workers exceeded the reference value of 1×10^{-4}. By indication, exposure to PAHs has a relatively higher cancer risk than exposure to heavy metals. In the case of PAHs, the ILCR was 9.5×10^{-5} for hall workers and 1.0×10^{-4} for kitchen workers. The difference in the ILCR of the two groups was not statistically significant ($p>0.05$), and of the 25 workers, only two hall workers (8%) and one kitchen worker (4%) exceeded $< 1\times10^{-4}$. **(Figure 6).**

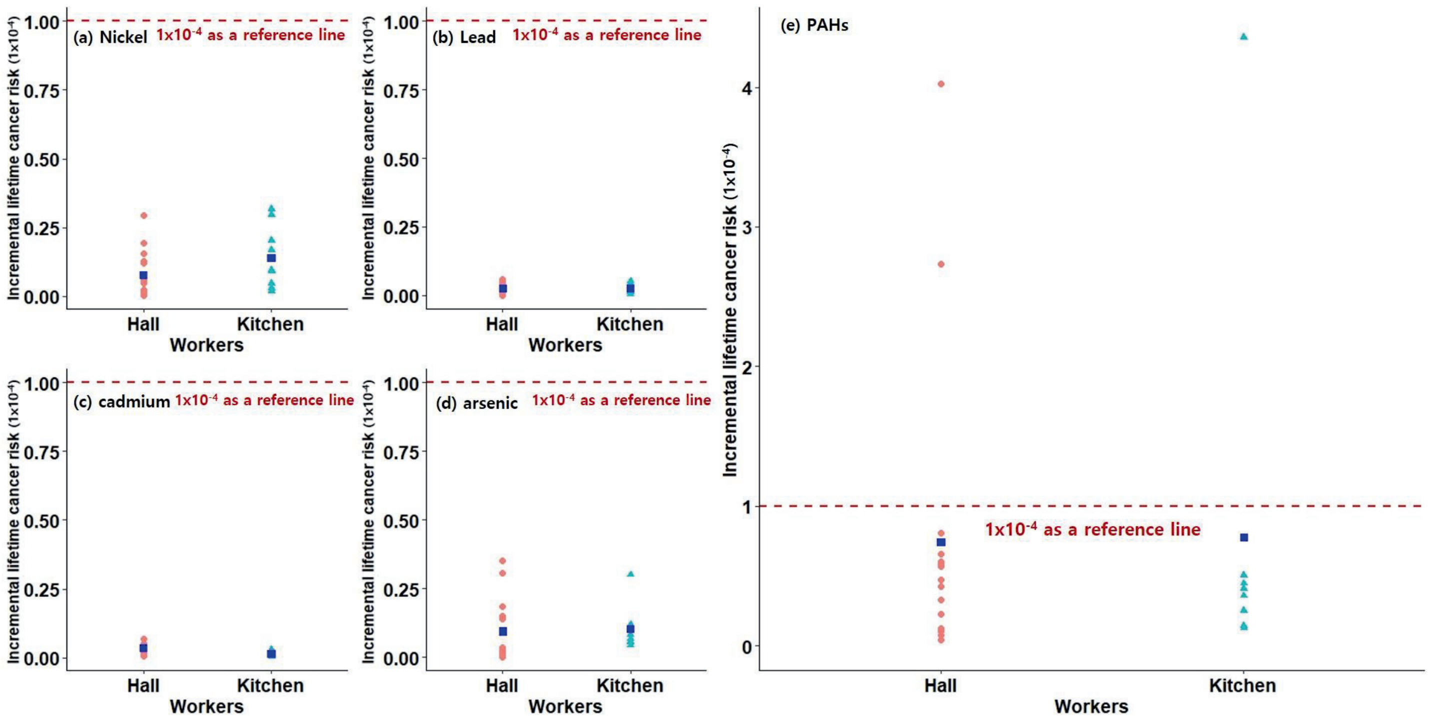

Figure 6. Incremental lifetime cancer risk estimation of workers based on concentrations of PAHs and heavy metals (; nickel, lead, cadmium and arsenic) in three Korean BBQ restaurants. Value shown is mean (closed blue square).

4. Discussion

To our knowledge, this is the first study to evaluate exposure of workers in Korean BBQ restaurants to TSP, heavy metals, and PAHs, as well as the first study to derive the corresponding cancer risks. The GM concentration of TSP in the hall workers and kitchen workers was 141.45±2.08 μg/m^3 and 41.45±1.88 μg/m^3, respectively, and it was confirmed that the personal exposure value of hall workers handling charcoal was much greater. In the case of heavy metals, the exposure values of both hall and kitchen workers were below Korea OEL. However, the exposure values exceeded Korea's air quality standards for lead and the WHO's recommended standards for cadmium and manganese. In the case of PAHs, the exposure concentration of hall workers was 41.45±1.88 μg/m^3, which was higher than that of kitchen workers, 1.77±1.89μg/m^3, and it was confirmed that hall workers were more likely to be exposed to PAHs than kitchen workers. The cancer risk for heavy metals based on nickel, lead, cadmium, and arsenic did not exceed the reference values. However, in the case of 16 PAHs, among 25 workers, two hall workers and one kitchen worker exceeded the reference value, indicating that hall workers are more likely to have a greater cancer risk than kitchen workers.

The primary cooking method employed by the Korean BBQ restaurants in this study was charcoal. Several studies have shown that charcoal cooking produced more pollutants than other cooking methods. For example, a study that investigated night market stalls showed the highest PAHs concentrations (43.145 μg/m^3) in the stalls using the barbecue method (Zhao, et al., 2011). In the Taiwanese study, PAHs concentrations in Western fast food and Chinese cafeteria restaurants using roasting and frying were at the levels of 3.32 μg/m^3 and 3.99 μg/m^3, respectively, with lower concentrations than restaurants using charcoal (34.89μg/m^3) (Wu et al,.2019). Another study also emphasized that more particulate matter is generated in the barbecue process. This study reported that the TSP concentration when the charcoal itself was burned was 152.29 μg/m^3, and the TSP concentration generated during direct roasting was 37480.13 μg/m^3. Compared to charcoal self-burning, a significantly higher concentration occurred in the direct roasting process (Ahn et al., 2016).

In **Table 4**, the TSP level of the hall in Restaurant C (41.2±1.9 μg/m^3) was lower than the other two restaurants, and the reasons are as follows. In Restaurants A and B, a large amount of smoke was generated in the process of igniting the charcoal in the charcoal storage and bringing it to the table in the hall. However, in the case of Restaurant C, the charcoal was ignited directly at the table in the hall, following which the lid was closed and the ventilation hood placed as close as possible to minimize smoke. Previous studies demonstrated that running the exhaust fan after cooking for more than 15 minutes would reduce to $PM_{2.5}$ concentration, suggesting that better ventilation could help reduce exposure (Dobbin et al., 2018).

The GM concentrations of TSP in the kitchens were 92.63 ± 14.55 μg/m^3, 17.77 ± 11.64 μg/m^3 and 104.41 ± 70.02 μg/m^3 for Restaurants A, B, and C, respectively. In the kitchen, despite cooking continuously, such as boiling or roasting, the concentration was lower than the values of hall and charcoal storage. The $PM_{2.5}$ concentrations generated during roasting and boiling were 1107±340 μg/m^3 and 257±29 μg/m^3, respectively, which were 2.5 ~ 62.3 times higher than those in this study (Li et al., 2018). It is expected that the concentration in the kitchen in this study was lowered because the ventilation facilities were continuously operated in the process of cooking, and wet cleaning was performed once or twice every 2 hours.

In **Figure 2**, the hall worker ignites the charcoal in the charcoal storage and waits for 3 minutes, the ignition time, and moves the prepared charcoal to the hall. The hall worker, who had the highest exposure concentration (578.73 μg/m^3), continued to ignite the charcoal by staying in the charcoal storage (932.68 μg/m^3) because there were many restaurant users at the time (about twice the average sales).

Although the personal exposure values of heavy metals investigated in this study were lower than that of Korea's OEL, some heavy metals (lead, manganese, and cadmium) exceeded Korea's Air quality standards and the WHO's recommendations. The average patronage of the Korean BBQ restaurants in this study was 70 to 80 people a day, so these exposure values are a concern not only for the health of the workers, but also for the health of the visitors. As such, it is necessary to generate a plan to mitigate indoor pollution in restaurants by using these results as a basis for improving indoor air quality.

Table 8 shows the concentrations of PAHs and associated cancer risks from previous studies. As can be observed, PAH levels for street food cart workers was 8.79±1.6 μg/m^3. Compared to this value, the PAH exposure levels for the hall and kitchen workers in Korean grilled meat were 5.0 and 38.2 times greater, respectively. Relative to the restaurant workers, the four workers at the night market food stand had PAH exposures of 25.51 μg/m^3, 23.40 μg/m^3, 44.17 μg/m^3, and 28.37 μg/m^3, respectively. These values range from 40 to 76 times greater than those of restaurant workers. Despite food cart and night market vendors using the same charcoal ignition sources as the restaurants, their appears to be a drastic difference in the PAH concentrations. This can be attributed to the fact that, unlike the restaurants, cart workers did not have access to suitable ventilation facilities due to legal restrictions as well as lack of space in areas where they were set up for business. The PAH levels in this study are similar to workers in Taiwan.

Coke plant side oven workers had average PAH exposures of 0.99±1.05 μg/m^3, which is not significantly different from PAH exposures measured in this study. However, the PAHs measured in the restaurants were predominantly, 2-ring, 3-ring PAHs, whereas coke plant workers were exposed to 5-ring and 6-ring PAHs which have a higher potency and toxicity. For that reason, the cancer risk in coke ovens would be higher than that of this study.

This study has several limitations. Firstly, it is difficult to extrapolate and apply the results of this study to the general majority of Korean meat-grilling restaurants and associated workers. The restaurants analyzed in this study had average daily sales of approximately 1.5 million won per, and an average establishment size of 184.06 m^2 (medium-sized restaurants). Furthermore, only 3 workers were measured and analyzed per day for each restaurant. For a comprehensive study of PAH and heavy metal exposure in Korean BBQ restaurants, as well as generating estimates of cancer risks in populations with high exposure to those substances, future studies should include analyses of larger restaurants. Such restaurants may differ in terms of the number of workers as well as the and type of cooking methods utilized. Second, during the course of this study, the current Covid-19 pandemic was just beginning to emerge. Consequently, restaurant sales and patronage were approximately half of what they normally were. Third, the small sample size precluded assessment of other contributing factors, such as smoking in exposure to PAH and heavy metals. The small sample size also precluded assessment of gender-specific exposure and cancer risk. Fourth, given that only a few studies have calculated ILCR for air exposure of heavy metals based on cooking methods and for cooking-related workplaces, the current study could not include a comparative analysis of the cancer risks of heavy metals.

This study is the first to estimate the cancer risk of personal exposure to both PAHs and heavy metals in Korean BBQ restaurants. The estimation is more representative of personal exposure compared to previous studies that have investigated the characteristics and concentration of air pollutants according to the difference in the roasting method for cooking meat. This study investigated occupational exposure to substances-related cooking, and correlating ILCR of workers in the restaurants. Since cooking with charcoal is used in many countries other than Korea, this study could be used to provide information for controlling occupational exposures elsewhere.

Table 8 Comparison of total PAH concentrations (μg/m^3) and cancer risk in this study with other available data.

Emission source	City	Sampling location/subject		Measured PAHs	Total PAH (GM, μg/m^3)	BaP_{eq}	Reference
Cooking	Seoul, Korea	Korean grilled meat restaurant.	Hall workers	16 priority control PAHs	1.77±1.89	9.5×10^{-5}	This study
			Kitchen workers		0.23±2.04	1.0×10^{-4}	
	Kaohsiung, Taiwan	Commercial kitchens	Western fast food kitchen	16 priority control PAHs	3.17±1.34	0.22×10^{-4}	Wu et al.
			Chinese cafeteria kitchen		3.72±1.6	1.27×10^{-4}	
			Street food cart		8.79±6.1	1.15×10^{-4}	
	Taipei, Taiwan	Workers of food stands at night markets		16 priority control PAHs	25.51	1.01×10^{-4}	Zhao et al.
					23.40	1.90×10^{-4}	
					44.17	1.13×10^{-4}	
					28.37	1.03×10^{-4}	

		Bus line		0.11±0.04	6.98×10^{-6}	
Vehicle Inspection factory	China, Beijing	Gasoline line	16 priority control PAHs	0.06±0.02	7.05×10^{-6}	Li et al.
		Diesel line		0.20±0.11	1.28×10^{-5}	
Traffic	China, Tianjin	Traffic policeman	16 priority control PAHs	5.05	1.06×10^{-4}	Hu Y et al.
		Side-oven workers in coke plants		0.99±1.05	-	
Coking	Kaohsiung, Taiwan		16 priority control PAHs			Lin et al.
		Top-oven workers in coke plants		6.95±1.45	-	

5. Conclusion

This study evaluated the occupational exposure of TSP, PAHs, and heavy metals to workers in Korean grilled meat restaurants. The estimation was more representative of human exposure compared to previous studies that have used stationary samples to assess the carcinogenic potencies of cooking emissions and human exposure.

Hall workers igniting and carrying charcoal indicated higher TSP and PAHs exposure values than kitchen workers. Accordingly, more hall workers exceeded the reference value for cancer risk compared to kitchen workers.

The heavy metal exposure values of both hall and kitchen workers were below Korean occupational exposure limits. However, it exceeded Korea's air quality standards for lead and the WHO's recommended standards for cadmium and manganese.

This study not only provides data on the occupational exposure risks of Korean meat grilled restaurants, but can also be used to extrapolate risks of restaurant patrons, and suggests methods to mitigate these risks.

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국문초록

한국 육류직화구이 음식점 근로자의 입자상 물질, 중금속 및 다환방향족탄화수소 노출 평가

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연구배경: 한국 육류직화구이 음식점은 주로 숯을 연료로 사용하며 이를 이용하여 육류를 조리하는 과정에서 건강상 악영향을 미치는 입자상 물질, 중금속 및 다환방향족탄화수소와 같은 유해물질을 발생시킨다. 선행연구는 이러한 물질의 배출 특성을 연구한 것이 대부분이며, 근로자에 대한 개인 노출 평가를 조사한 연구는 거의 없는 실정이다. 따라서, 본 연구의 목적은 한국 육류직화구이 음식점에서 근무하는 종사자들에 대하여 입자상 물질, 중금속 및 다환방향족탄화수소와 같은 주요 유해 물질에 대한 직업 노출을 평가하고, 이에 상응하는 발암 위해도를 조사하는 것이다.

연구방법: 수도권 소재의 육류직화구이 음식점 3 곳을 선정하여 연구를 진행하였다. 홀, 부엌 및 숯 저장소와 음식점의 홀 및 근로자를 대상으로 총 분진, 시중 일부 숯 제품이 함유하고 있다고 알려진 중금속 6종 및 EPA에서 우선지정관리물질로 지정한 16 종의 PAHs에 대한 노출 평가를 진행하였다. 개인 및 지역 시료 모두 근로자의 호흡기 영역에서 휴식시간을 제외한 8시간동안 수집되었으며, 각 음식점마다 3일 반복 측정되었다. 총 분진의 경우 밸런스를 이용하여 정량적으로 무게를 재었으며, PAHs와 중금속의 경우 NIOSH method 5515와 7730을 기반으로 하여금 각각 GC-MS 와 ICP-MS를 이용하여 정량분석을 실시하였다. 발암 위해도는 US EPA의 Risk Assessment Guidance에 따라 계산 및 평가하였다.

결과: 장소 별 총 분진 평균 농도는 숯 저장소 > 홀 > 부엌 > 대조군(외기) 순서인 것을 나타냈다. 홀 근로자의 총 분진 농도 141.45±2.08 μg/m^3 수준으로 주방 근로자(41.15±1.88 μg/m^3)보다 약 3.8 배 높음을 확인하였다(p <0.001). 본 연구에서 측정 한 중금속의 지역 및 개인 시료에서 둘다 한국의 OEL 미만이었으나, 납의 경우 대기환경기준을, 카드뮴 및 망간의 경우 WHO 권고기준을을 초과하였다. 총 PAHs의 평균 농도는 홀, 부엌 및 숯 저장에서 통계적으로 차이가 나지 않았으며, (각각 0.21± 4.81 μg/m^3, 0.30 ±2.69 μg/m^3, 0.27 ±4.66 μg/m^3). 홀 근로자 및 주방 근로자에 대한 총 PAHs의 평균 농도는 각각 1.77±1.89 μg/m^3 및 0.23±2.04 μg/m^{3}으로, 숯을 직접적으로 다루는 홀 근로자가 주방근로자보다 더 노출되는 것을 확인하였다. PAHs의 발암 위해도는 홀 근로자의

경우 9.5×10^{-5}, 주방 근로자의 경우 1.0×10^{-4}이였으며, 25명의 근로자 중 2명의 홀 근로자와 1명의 주방 근로자가 기준을 초과하였다.

결론: 본 연구는 한국 육류직화구이 음식점의 근로자들에 대에 총분진, 중금속, 다환방향족탄화수소를 노출 평가하였으며, 그에 따르는 발암 위해도를 평가하였다. 숯을 점화 및 운반하는 홀 근로자는 주방 근로자보다 높은 TSP 및 PAHs 노출 값을 나타내었으며, 더 많은 홀 근로자가 주방 근로자보다 발암위험기준을 초과하였다. 개인의 중금속 노출 수준은 한국의 직업 노출 한계 미만이었다. 그러나 납의 경우 한국 대기환경기준을 초과하였으며, 카드뮴 및 납은 WHO의 권고 기준을 초과하였다. 한국 육류직화구이 음식점은 관련 근로자뿐만 아니라, 다양한 연령대의 이용객이 이용하므로 이러한 유해물질은 가능한 낮은 수준으로 관리될 필요가 있다.

다환방향족탄화수소, 중금속, ICLR, 노출 평가,
육류직화구이

학번: 2018 - 23627

Appendices

Table A-1. Formula and physical properties of EPA 16 PAHs and carcinogen classification

Name	Molecular Formula	Molecular Weight	Boiling Point	IARC
Naphthalene	$C_{10}H_8$	128.18	218	2B[a]
Acenaphthylene	$C_{12}H_8$	152.20	265-280	2B
Acenaphtene	$C_{12}H_{10}$	154.20	277-279	3[b]
Fluorene	$C_{13}H_{10}$	166.23	293-295	3
Anthracene	$C_{14}H_{10}$	178.24	340	3
Phenanthrene	$C_{124}H_{10}$	178.24	339-340	3
Fluoranthene	$C_{16}H_{10}$	202.26	375-393	3
Pyrene	$C_{16}H_{10}$	202.26	360-404	3
Benzo[a]anthracene	$C_{18}H_{12}$	228.30	435	2B
Chrysene	$C_{18}H_{12}$	228.30	441-448	2B
Benzo[b]fluoranthene	$C_{20}H_{12}$	252.32	481	2B
Benzo[k]fluoranthene	$C_{20}H_{12}$	252.32	480-481	2B
Benzo[a]pyrene	$C_{20}H_{12}$	252.32	493-496	1[c]
Benzo[g,h,i]perylene	$C_{22}H_{12}$	276.34	524	3
Indeno[1,2,3-c,d]pyrene	$C_{22}H_{12}$	276.34	-	2B
Dibenzo[a,h]anthracene	$C_{22}H_{14}$	278.35	525	2A[d]

Table A-2. The results of quality control for PAHs

Chemicals/species	RT	Recovery rate(%)		Coefficient of variation for analysis	Calibration line parameter			LOD(µg/m³)	
	(Min)	Filter	XAD-2	(%)	Slope	R^2	R	Filter	XAD-2
Naphthalene	9.57	1.6%	47.4%	2.45%	0.298	0.9955	0.9978	0.164	0.007
Acenaphthylene	13.14	4.5%	53.4%	2.89%	0.867	0.9984	0.9992	0.025	0.003
Acenaphthene	13.58	12.3%	61.1%	4.93%	0.607	0.9975	0.9987	0.008	0.002
Fluorene	14.77	16.3%	66.5%	2.88%	0.606	0.9979	0.9989	0.008	0.003
Phenanthrene	17.16	29.7%	62.4%	3.37%	0.645	0.9994	0.9997	0.003	0.002
Anthracene	17.29	42.9%	66.6%	5.16%	0.460	0.9971	0.9985	0.003	0.003
Fluoranthene	21.07	50.1%	60.0%	8.58%	0.354	0.9977	0.9989	0.005	0.006
Pyrene	21.87	50.2%	58.0%	9.27%	0.351	0.9965	0.9982	0.005	0.005
Benzo(a)anthracene	26.82	83.7%	74.4%	4.02%	1.783	0.9986	0.9993	0.002	0.003
Chrysene	26.98	80.5%	64.1%	5.10%	1.883	0.9986	0.9993	0.005	0.008
Benzo(b)fluoranthene	31.35	80.9%	53.3%	8.21%	12.411	0.9966	0.9983	0.025	0.049
Benzo(k)fluoranthene	31.45	67.4%	53.1%	9.58%	13.012	0.9969	0.9985	0.026	0.042

Benzo(a)pyrene	32.56	85.6%	62.2%	10.27%	8.868	0.9982	0.9991	0.015	0.026
Indeno(1,2,3-cd)pyrene	36.68	101.1%	63.5%	15.98%	6.978	0.9970	0.9985	0.008	0.016
Dibenz(a,h)anthracene	36.78	73.4%	54.6%	15.85%	3.258	0.9985	0.9992	0.009	0.015
Benzo(g,h,i)perylene	37.51	86.6%	52.4%	13.16%	8.388	0.9976	0.9988	0.011	0.023

Table A-3. TSP concentration in air (μg/㎥) determined via personal and sampling in three Korean BBQ restaurants

Location	N	AM± SD	GM(GSD)	Range
Personal measurement				
Hall	16	181.3±139.1	141.45(2.08)	46.33-578.73
Kitchen	7	47.9±24.2	41.15(1.88)	12.35-81.36
Total	25	133.25±128.72	90.69(2.49)	12.35-578.73
Area measurement				
Hall	9	97.7±43.3	86.61(1.76)	33.23-161.15
Kitchen	9	62.0±45.2	44.39(2.60)	10.50-137.25
Storage	9	256.7±262.1	196.30(1.97)	107.38-932.68
Outdoor	9	21.6±17.6	15.37(2.47)	5.24-53.77
Total[a]	27	138.80±172.56	91.05(2.59)	10.50-932.68

[a] The outdoor value is excluded.

Table A-4. Heavy metals concentration in air (μg/㎥) determined via personal sampling in three Korean BBQ restaurants

Species /Chemical	Hall(N=16)			Kitchen(N=9)			Total(N=25)		
	Mean (SD)	GM (GSD)	Range	Mean (SD)	GM (GSD)	Range	Mean (SD)	GM (GSD)	Range
Cr	8.16(4.54)	7.39(1.54)	3.98-23.37	7.19(1.92)	6.96(1.31)	4.66-9.92	7.81(3.79)	7.23(1.46)	3.98-23.37
Mn	13.38(27.79)	3.31(5.49)	<LOD-98.05	1.19(0.82)	0.97(2.06)	<LOD-2.10	9.80(23.75)	2.31(4.79)	<LOD-98.05
Ni	2.00(2.27)	1.49(2.02)	0.49-0.06	2.52(2.92)	1.76(2.24)	0.61-10.08	2.19(2.48)	1.59(2.07)	0.49-10.22
As	0.12(0.08)	0.10(2.03)	0.02-0.33	0.12(0.10)	0.09(2.07)	0.04-0.31	0.12(0.08)	0.09(2.02)	0.02-0.33
Cd	0.04(0.02)	0.04(1.55)	<LOD-0.07	0.03(0.01)	0.02(1.37)	<LOD-0.04	0.03(0.02)	0.03(1.53)	<LOD-0.07
Ba	2.60(2.08)	1.99(2.15)	0.57-8.74	1.41(0.52)	1.33(1.42)	0.88-2.45	2.17(1.77)	1.72(1.95)	0.57-8.74
Pb	1.07(1.18)	0.78(2.17)	0.24-5.27	1.13(1.01)	0.89(2.00)	0.25-3.73	1.09(1.10)	0.82(2.08)	0.24-5.27

Table A-5. Heavy metals concentration in air (μg/㎥) determined via area sampling in three Korean BBQ restaurants

Species /Chemical	Hall(N=9)			Kitchen(N=9)			Storage(N=8)			Outdoor(N=9)		
	Mean (SD)	GM (GSD)	Range	Mean (SD)	GM (GSD)	Range	Mean (SD)	GM (GSD)	Range	Mean (SD)	GM (GSD)	Range
Cr	9.1 (2.2)	8.90 (1.29)	5.48-12.30	8.5(1.6)	8.36 (1.20)	<LOD - 11.61	8.4(3.8)	7.72 (1.53)	4.25-15.95	7.4 (1.7)	7.21 (1.28)	4.66-9.89
Mn	1.3 (1.0)	1.05 (2.09)	<LOD-2.72	1.8(1.0)	1.55 (1.96)	<LOD - 2.94	83.9 (189.4)	7.65 (12.07)	<LOD - 470.38	1.1 (0.5)	0.98 (1.54)	<LOD - 1.73
Ni	14.1 (31.5)	4.85 (3.29)	1.61-97.96	4.2(6.9)	2.16 (2.97)	<LOD - 21.16	3.1(4.8)	1.80 (2.51)	0.70-14.81	1.8 (1.2)	1.56 (1.86)	0.50-4.54
As	0.2 (0.1)	0.12 (2.22)	0.03-0.34	0.1(0.1)	0.08 (2.34)	<LOD - 0.24	0.1 (0.0)	0.08 (1.47)	0.04-0.13	0.1 (0.1)	0.08 (1.66)	0.05-0.23
Cd	0.2 (0.3)	0.07 (4.42)	LOD-0.77	0.1(0.1)	0.07 (2.38)	<LOD-0.18	0.0 (0.0)	0.02 (1.37)	<LOD - 0.04	0.1 (0.1)	0.09 (2.28)	<LOD - 0.16
Ba	2.5 (1.5)	2.09 (1.97)	0.71-4.99	2.2 (1.0)	1.94 (1.78)	<LOD-3.43	4.2 (5.0)	2.59 (2.73)	0.76-15.87	1.8 (1.8)	1.40 (2.01)	0.68-6.41
Pb	1.8 (1.7)	1.29 (2.28)	0.4-5.4	1.1 (0.5)	0.94 (1.72)	<LOD-1.49	1.3 (1.0)	0.96 (2.42)	0.19-3.11	0.8 (0.3)	0.78 (1.56)	0.30-1.44

Table A-6. Incremental lifetime cancer risk($\times 10^{-4}$) estimation of personal heavy metals in three Restaurant by location in three Korean BBQ restaurants.

Restaurant	Location		Species/Chemical			
			Ni	Pb	Cd	As
A	Hall(N=6)	Mean±SD	0.054±0.053	0.024±0.026	0.027±0.027	0.113±0.113
		GM(GSD)	0.038(2.46)	0.013(3.35)	0.024(1.71)	0.053(4.08)
		Range	0.012-0.156	0.003-0.059	0.011-0.047	0.013-0.351
	Kitchen(N=3)	Mean±SD	0.136±0.051	0.041±0.008	0.051±0.020	0.170±0.126
		GM(GSD)	0.130(1.44)	0.040(1.22)	0.047(1.57)_	0.136(2.36)
		Range	0.095-0.194	0.033-0.049	0.029-0.070	0.055-0.305
B	Hall(N=6)	Mean±SD	0.118±0.106	0.029±0.019	0.008±0.001	0.124±0.108
		GM(GSD)	0.087(2.25)	0.024(1.84)	0.008(1.12)	0.086(2.65)
		Range	0.049-0.294	0.014-0.056	0.008-0.009	0.029-0.301
	Kitchen(N=3)	Mean±SD	0.248±0.103	0.049±0.007	0.044±0.00	0.115±0.030
		GM(GSD)	0.230(1.65)	0.049(1.17)	0.044(0.00)	0.112(1.32)
		Range	0.129-0.319	0.041-0.054	<LOD-0.044	0.083-0.141
C	Hall(N=4)	Mean±SD	0.033±0.040	0.003±0.002	0.004±0.00	0.005±0.002
		GM(GSD)	0.019(3.35)	0.002(2.45)	0.004(0.00)	0.005(1.60)

	Range	0.005-0.092	0.001-0.006	<LOD-0.004	0.003-0.007
	Mean±SD	0.068±0.088	0.012±0.015	0.007±0.00	0.031±0.032
Kitchen(N=3)	GM(GSD)	0.035(4.18)	0.007(3.88)	0.007(0.00)	0.021(2.96)
	Range	0.010-0.169	0.002-0.029	<LOD-0.007	0.008-0.068
	Mean±SD	0.101±0.096	0.025±0.021	0.027±0.021	0.098±0.105
Total(N=25)	GM(GSD)	0.059(3.18)	0.014(3.69)	0.020(2.48)	0.049(3.96)
	Range	0.005-0.319	0.001-0.059	<LOD-0.004	0.003-0.351

Table A-7. Incremental lifetime cancer risk(×10^{-4}) estimation of personal PAHs in three Restaurant by location in three Korean BBQ restaurants.

Restaurant	Location		Species/Chemical							
			NAP	ACY	ACE	FLU	PHE	ANT	FLUR	PYR
A	Hall (N=6)	Mean ±SD	0.459±0.629	0.202±0.315	0.090±0.147	0.144±0.175	0.112±0.112	0.008±0.006	0.005±0.005	0.040±0.074
		GM (GSD)	0.278(2.61)	0.108(2.81)	0.033(4.49)	0.096(2.43)	0.082(2.30)	0.006(2.01)	0.004(2.00)	0.012(4.80)
		Range	0.129-1.736	0.053-0.845	0.008-0.384	0.035-0.499	0.025-0.336	<LOD-0.017	0.002-0.016	0.004-0.190
	Kitchen (N=3)	Mean ±SD	0.164±0.138	0.093±0.040	0.097±0.098	0.067±0.007	0.064±0.007	<LOD	<LOD	<LOD
		GM (GSD)	0.130(2.25)	0.087(1.58)	0.068(3.51)	0.067(1.12)	0.064(1.11)	<LOD	<LOD	<LOD
		Range	0.067-0.321	0.053-0.133	<LOD-0.166	0.059-0.073	0.059-0.072	<LOD	<LOD	<LOD
B	Hall (N=6)	Mean ±SD	0.619±0.717	0.266±0.318	0.141±0.164	0.192±0.164	0.204±0.290	0.043±0.007	0.015±0.014	0.068±0.113
		GM (GSD)	0.239(5.96)	0.158(2.95)	0.078(3.26)	0.147(2.16)	0.081(5.21)	0.042(1.18)	0.011(2.48)	0.017(7.31)
		Range	0.020-1.711	0.047-0.871	0.025-0.416	0.072-0.476	0.009-0.782	0.038-0.047	0.005-0.035	0.003-0.237

	Kitchen (N=3)	Mean ±SD	0.223±0.090	0.089±0.00	0.102±0.076	0.079±0.00	0.158±0.145	<LOD	<LOD	<LOD
		GM (GSD)	0.212(1.49)	0.089(0.00)	0.086(2.28)	0.079(0.00)	0.108(3.18)	<LOD	<LOD	<LOD
		Range	0.145-0.322	LOD-0.089	0.048-0.155	LOD-0.079	0.032-0.317	<LOD	<LOD	<LOD
C	**Hall (N=6)**	Mean ±SD	0.022±0.014	0.019±0.009	0.017±0.005	0.019±0.006	0.015±0.010	0.001±0.00	0.002±0.00	0.001±0.00
		GM (GSD)	0.019(1.90)	0.016(2.03)	0.016(1.41)	0.018(1.38)	0.011(3.21)	0.001(0.00)	0.002(0.00)	0.001(0.00)
		Range	0.010-0.041	0.006-0.025	0.011-0.022	0.012-0.024	0.002-0.026	<LOD-0.001	<LOD-0.002	<LOD-0.001
	Kitchen (N=3)	Mean ±SD	0.027±0.024	0.035±0.010	0.020±.0016	0.017±0.006	0.021±0.018	<LOD	<LOD	<LOD
		GM (GSD)	0.021(2.28)	0.034(1.39)	0.017(2.17)	0.016(1.37)	0.012(4.42)	<LOD	<LOD	<LOD
		Range	0.013-0.055	0.023-0.041	0.008-0.038	0.013-0.023	0.002-0.039	<LOD	<LOD	<LOD
Total(N=25)		Mean ±SD	0.312±0.499	0.146±0.236	0.083±0.119	0.105±0.135	0.107±0.165	0.017±0.019	0.009±0.010	0.047±0.084
		GM (GSD)	0.113(4.58)	0.071(3.15)	0.039(3.35)	0.060(2.87)	0.047(4.15)	0.008(3.83)	0.005(2.53)	0.011(5.73)
		Range	0.010-1.736	<LOD-0.871	<LOD-0.416	<LOD-0.499	<LOD-0.782	<LOD-0.047	<LOD-0.035	<LOD-0.237