Charcoal Grill Restaurants Deteriorate Outdoor Air Quality by Emitting Volatile Organic Compounds

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Abstract

Our study investigated outdoor air pollution caused by volatile organic compounds (VOCs) such as BTEX and *n*-alkanes emitted from charcoal grill restaurants. Twenty sites were selected from a city in Korea for outdoor-air sampling. On average, the concentrations of all VOCs in the areas of the charcoal grill restaurants were significantly 1.3-to-2.6-fold higher than those in the non-charcoal-grill areas at the 1% level. The results showed that charcoal grill restaurants are responsible for both indoor and outdoor air pollution, which can cause health problems for pedestrians and residents in such areas as well as restaurant workers and customers.

Keywords: *n*-alkanes, BTEX, charcoal grill restaurant, outdoor air pollution, VOCs

Introduction

Charcoal has been extensively used in Korea for the grilling of food, especially meats such as pork and beef, and poultry such as chicken and duck. This cooking style, the meats and poultry prepared, and the distinctive flavors produced have all grown in popularity. A recent survey showed that fully 91.0% of Koreans prefer charcoal grilling and barbecuing over other food preparation methods, and that 71.3% favor pork among meats and poultry [1].

However, the use of charcoal for grilling meat has been reported to produce various kinds of hazardous chemicals, including heavy metals, polycyclic aromatic hydrocarbons (PAHs), heterocyclic aromatic amines (HCAs), BTEX (benzene, toluene, ethylbenzene, and *o,m,p*-xylenes), alkanes, alkenes, aldehydes, ketones, carboxylic acids, and others [2, 3]. Many of these compounds, such as benzene, PAHs, and HCAs, are known or suspected to be carcinogenic to humans [4]. Restaurant customers are exposed to

them *via* two major routes during their stay (~ 2 h per visit): oral ingestion of grilled meat and inhalation of polluted indoor air. Obviously, restaurant workers are even more highly exposed *via* inhalation over the course of long working hours (maximum ~ 12 h per day). The chemicals are released or produced from both charcoal itself [5, 6] and the food cooked, specifically as a result of the application of high temperature and subsequent Maillard reactions [7, 8].

Surprisingly, most people are unconcerned about these hazards, preferring to enjoy the pleasant aroma and taste of food so prepared, even at the cost of adverse health effects. And currently no local, regional, or national government agencies are intervening with any regulatory proposals or actions; either they do not consider the problems to be serious, or do not yet have sufficient data to take action.

Pollution of the air in areas surrounding grill restaurants is an additional problem, as the exhaust chimneys of such buildings typically are not equipped with any pollution-control devices. To date, however, no detailed data on the quality of the outdoor air in the vicinities of these restaurants has been reported.

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Chuncheon, the study area of the present research, has a population of 275,655 (as of Dec. 31, 2011). It is estimated that this city has more than 150 charcoal grill restaurants wherein pork, beef, chicken, duck, and other meats and poultry are cooked, most intensively during the dinner time. An aggravating factor during these evening hours is the radiation inversion layer that forms near ground level and which prevents complete diffusion of gaseous- and particulate-phase pollutants, thereby increasing their concentrations.

The aim of this study was to investigate the deterioration of outdoor air quality by charcoal grill restaurants. A total of 20 sites comprising two groups of 10 sites each were visited three times to determine the concentrations of VOCs such as BTEX and *n*-alkanes, on which basis the two groups were compared.

Experimental Procedures

Chemicals

Benzene, toluene, ethylbenzene, *p*-xylene, and *o*-xylene were purchased individually from Sigma-Aldrich and each mixed together in methanol to a concentration of 2,000 $\mu g \cdot mL^{-1}$. A mixture standard (1,000 $\mu g \cdot mL^{-1}$) of C_7-C_{30} *n*-alkanes was purchased from Supelco; five alkanes only (*n*-heptane, *n*-octane, *n*-nonane, *n*-decane, and *n*-undecane) were included in a quantitative determination.

Outdoor-Air Sampling

The outdoor-air sampling sites were categorized into A and B groups according to the number of charcoal grill restaurants within a specified area (Fig. 1). Ten group A sites (designated as •) had no charcoal grill restaurant within a radius of 30 m; and 10 group B sites (designated as ♦) had at least three such restaurants within the same distance. At least one A-B pair of sites in relatively close proximity to each other (within ~ 60 m) but differing markedly in the type of restaurant was visited on the same day for the purposes of inter-group comparison.

Air sampling was conducted for two site pairs simultaneously (four samples per day) for 1 h between 7 p.m. and 8 p.m. from Oct. 11 to Nov. 7, 2011. Each of four staff members visited an assigned site, walking within the specified area with a portable pump (210 series, SKC, Inc.) affixed at the waist and an adsorbent trap at the shoulder. Sampling at each site was repeated three times; a total of 60 air samples were collected. Sampling was conducted only on weekdays due to the probable concentration variability between weekdays and weekends. During the visits air temperature, wind speed, and relative humidity ranged from 7.0 to 15.5ºC (mean 12.16ºC), from 0.5 to 1.0 m·s⁻¹ (mean 0.79 m·s⁻¹), and from 55.1 to 89.4% (mean 73.9%), respectively. There was no rainfall during any of the samplings.

Each adsorbent trap was filled with 140 mg of Tenax® TA (1:1 ratio of 60/80 mesh and 20/35 mesh, Supelco, Inc.)
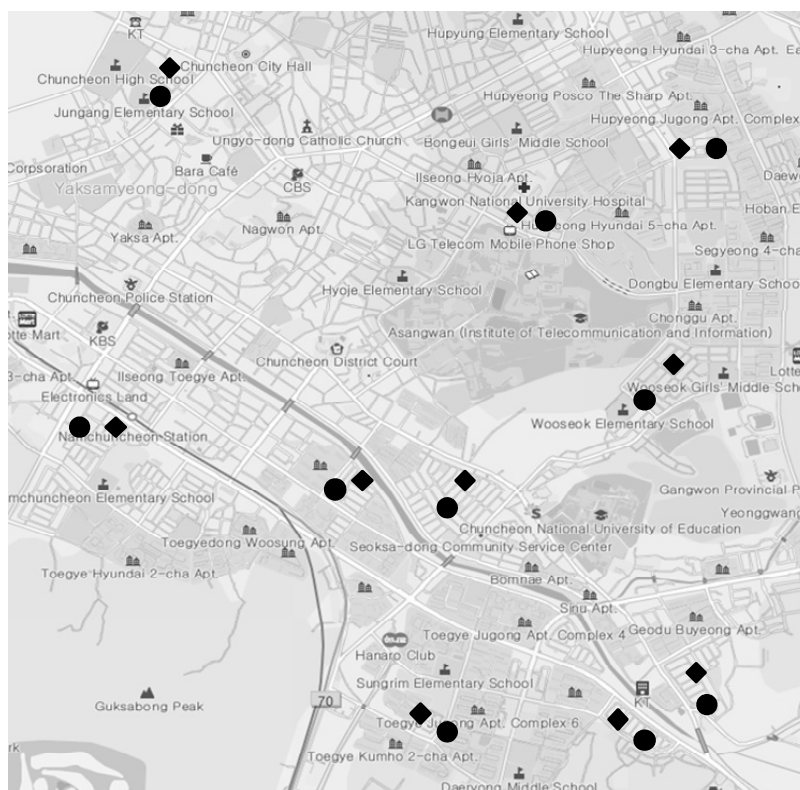

Fig. 1. Air sampling locations for measurement of airborne volatile organic compounds (VOCs) in Chuncheon, Korea. The study sites were categorized into two groups (distinguished by ♦ and •). The filled diamonds (♦) indicate the target sites where at least three charcoal grill restaurants are located within a radius of approximately 30 m, whereas the filled circles (•) represent sites where no charcoal grill restaurants are within the same distance.

and 80 mg of Carboxen 569 (20/45 mesh, Supelco, Inc.). The traps were cleaned at 300ºC for 3 h under He flow (50 mL·min⁻¹) prior to use. The air sampling was carried out at a flow rate of 200 mL·min⁻¹.

Sample Analysis

A cold trap containing 30 mg of Tenax® (60/80 mesh) was cooled to -30ºC to enrich the analytes, which had been thermally desorbed from the adsorbent trap at 280ºC for 6 min. The cold trap was then heated at 300ºC for 8 min to desorb the analytes, which were transferred to the gas chromatograph (GC) inlet through a capillary tube maintained at 150ºC, and finally injected into the gas chromatograph/mass spectrometer (GC/MS) at a split ratio of 20:1. The temperature of the GC injector was maintained at 250ºC. The analytical column was a capillary ZB-5 (30 m length × 0.25 mm i.d. × 0.5 µm film thickness, Phenomenex Inc.), through which He flowed at a rate of 1 mL·min⁻¹. The oven temperature was programmed as follows: remain at 32ºC for 10 min; increase to 250ºC at 5ºC min⁻¹; stay at 250ºC for 10 min. The transfer line between the GC and MS was maintained at 280ºC. The temperatures of the MS ion source and quadrupoles were 230 and 150ºC, respectively. Quantitative determination of the analytes was conducted in the selected ion monitoring (SIM) mode (m/z 77 and 78 for benzene, 77 and 91 for toluene, 77, 91, and 106 for ethylbenzene and xylenes, and 41, 43 for five *n*-alkanes).

Quality Assurance/Quality Control in Sampling and Analysis

To determine if there might occur any breakthrough problems with the sorbent traps during field sampling, two adsorbent traps were connected in series, one of which was attached to a pump that was operated at 200 mL·min⁻¹ for 1 h at one of the sampling sites. This test was carried out twice. In both cases, the analytes were found only in the front trap (not in the backup trap), indicating that no breakthrough problem was likely to be encountered during air sampling. Field sampling reproducibility, as tested by triplicate field sampling, showed a relative standard deviation (RSD) between 4.2 and 7.6% for the analytes, which confirmed the reliability of the sampling protocol.

Five-point (2, 10, 20, 50, and 80 ng) calibration curves were prepared for all analytes by adsorbing analytes (vaporized at 280ºC using a gas sampling bulb) onto traps. The coefficients of determination (r^2) of the linear curves for all the compounds were satisfactory, ranging from 0.9957 (for *n*-undecane) to 0.9996 (for *n*-nonane). During the sample analysis, an external standard trap (30 ng) was run every 10 samples to validate the calibration curves. Each external check showed a relative percent difference (RPD) of 20% or less between the expected and measured values.

Analytical reproducibility was tested by running seven traps, each containing 20 ng of each analyte; the results showed an RSD of 10% or less in all compounds. The method detection limits (MDLs) were estimated by running seven traps, each containing 0.2 ng of each analyte (0.4 ng for *n*-octane) and multiplying these estimated detection limits by 3. The MDLs for 1-h sampling were estimated by the following formula: 3.14 × standard deviation of seven replicates, or 0.01, 0.04, 0.05, 0.07, 0.06, 0.02, 0.08, 0.04, 0.07, and 0.08 µg·m⁻³ for benzene, toluene, ethylbenzene, *m,p*-xylenes, *o*-xylene, *n*-heptane, *n*-octane, *n*-nonane, *n*-decane, and *n*-undecane, respectively.

Data Analysis

Prior to statistical data analysis, outlying points, as determined mathematically, were excluded. Paired *t*-tests were conducted to find the VOC concentration differences between groups A and B. Inter-group correlation analyses also were performed. Excel (Microsoft Co.) and Systat® (Systat Software, Inc.) 12 were employed.

Results

Summary of VOC Concentrations

Fig. 2 shows a representative field-sample chromatogram. As is indicated, BTEX and the five *n*-alkanes included in the quantitative determination were found in the highest concentrations.

The concentrations of the BTEX and five *n*-alkane compounds measured in both groups were all above the MDLs, as shown in the Table 1 summary. The concentrations of toluene were the highest in groups A and B, with
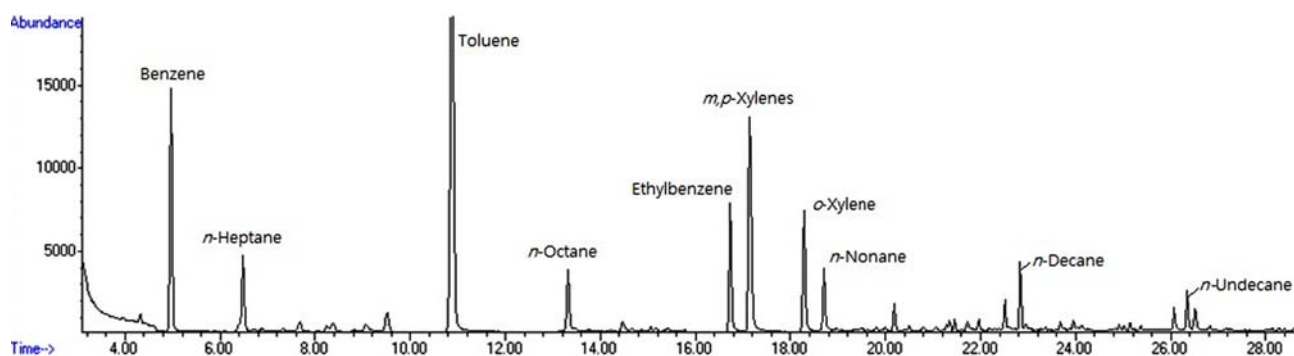

Fig. 2. Representative GC/MS chromatogram of an air sample from one target site.

Table 1. Summary of concentrations of BTEX and volatile alkanes measured for two groups of sites*

Descriptive statistic	Benzene		Toluene		Ethylbenzene		m,p-Xylenes		o-Xylene	
	Group A	Group B	Group A	Group B	Group A	Group B	Group A	Group B	Group A	Group B
Mean±sd	1.82±1.22	2.93±1.41	2.61±1.98	4.16±2.70	0.464±0.442	1.19±1.23	0.791±0.715	1.64±1.66	0.360±0.360	0.783±1.01
Median	1.40	3.00	2.08	4.08	0.372	0.560	0.633	1.15	0.281	0.475
Min-max	0.365-5.56	0.645-5.85	0.253-7.58	0.296-10.2	0.0640-2.03	0.0765-4.68	0.0813-3.15	0.194-6.27	0.0571-1.78	0.0981-5.26

Statistic	n-Heptane		n-Octane		n-Nonane		n-Decane		n-Undecane	
	Group A	Group B	Group A	Group B	Group A	Group B	Group A	Group B	Group A	Group B
Mean±sd	0.518±0.355	0.832±0.576	0.246±0.130	0.455±0.287	0.302±0.169	0.416±0.201	0.444±0.533	0.563±0.340	0.230±0.220	0.436±0.488
Median	0.437	0.611	0.232	0.412	0.244	0.371	0.282	0.395	0.171	0.260
Min-max	0.0881-1.17	0.158-2.51	0.0867-0.529	0.113-1.23	0.125-0.903	0.169-1.11	0.0733-3.02	0.192-1.27	0.00816-0.776	0.0187-2.45

*Paired-tests of groups A and B showed significant differences between them at the 1% level of significance (excepting n-decane).

means of 2.61 and 4.16 $\mu g \cdot m^{-3}$, respectively. Benzene was found in the next-highest concentrations, 1.82 and 2.93 $\mu g \cdot m^{-3}$ on average in groups A and B, respectively. The other eight compounds were present below 1 $\mu g \cdot m^{-3}$ in both groups, excepting ethylbenzene and the *m,p*-xylenes in group B.

Comparison of VOC Concentrations between Two Groups

The average concentration of each compound in group B was 1.3 (*n*-decane)-to-2.6 (ethylbenzene)-fold higher than in group A, suggesting that charcoal grilling indeed contributes to the elevation of each compound in areas surrounding restaurants (Table 1).

To identify any statistical differences in the average concentrations of each compound between the two groups, paired *t*-tests were carried out. One, 3, 4, 2, 2, 0, 4, 4, 4 and 2 pair(s) of data were eliminated in the analyses for benzene, toluene, ethylbenzene, *m,p*-xylenes, *o*-xylene, *n*-heptane, *n*-octane, *n*-nonane, *n*-decane, and *n*-undecane, respectively, since at least one value of each pair was found to be a mathematical outlier. The results of the paired *t*-tests showed that the concentrations of all of the compounds significantly differed between the two groups with $p < 0.01$, indicating once again that charcoal grilling at restaurants contributes to the elevation of outdoor VOC concentrations.

Fig. 3 shows box plots comparing the concentrations of total VOCs (a), total BTEX (b), and total alkanes (c) between groups A and B. As was the case for the individual compounds, the mean (solid line) and median values (dotted lines) in group B were significantly higher than those in group A at the 1% level.

Correlations between VOC Compounds

Correlation analyses were performed on the two groups' VOC concentrations, the results of which are shown in Table 2. In group A, benzene and toluene were not correlated with any other compounds, whereas the other compounds were significantly correlated with at least one (e.g., *n*-hexane) to five (e.g., *o*-xylene and *n*-octane) compounds at either the 1 or 5% level. In group B, no correlations of benzene with other compounds were found. The other compounds, however, were significantly correlated with at least a single (e.g., toluene) to five (e.g., *n*-nonane and *n*-decane) compounds at the 1 or 5% level. Notably, the correlation patterns (the number of significant correlations for each compound and for each set of compounds) in group B were quite different from those in group A.

Discussion

This research investigated and disclosed the deterioration of outdoor air quality effected by restaurants that charcoal-grill meat. Though this study was conducted in a city located in Korea, similar types of outdoor air pollution problems from charcoal grilling may take place in other

countries due to worldwide growing consumption of grilled meat (e.g., hamburgers, outdoor grilling) [9].

From the public health perspective, indoor air quality is more important than outdoor air quality, since most people spend more time indoors [10, 12], where, in charcoal grill restaurants for example, they can be directly exposed to high concentrations of VOCs. For this reason, recent research has focused on the source profiles of VOCs emitted during cooking processes [2, 3, 7, 8].

BTEX and five *n*-alkanes were selected as the foci of the present study, owing to the fact that they can act as either directly hazardous chemicals or as precursors of ozone formation; additionally, they were the major compounds detected on a representative field-sample gas chromatogram. Acute toxic effects such as skin irritation and CNS problems (e.g., tiredness, dizziness, headache, loss of coordination, etc.) are not to be expected from exposure to low levels of environmental BTEX; however, chronic exposure to benzene can cause cancers (known human carcinogens (Group A) such as leukemia [13]. Although *n*-alkanes are not as toxic as BTEX, they are involved in photochemical smog events through the oxidative formation of secondary organic aerosols [14].

In addition to BTEX and the five *n*-alkanes, more compounds were identified using NIST Mass Spectral Library with Search Program (ver. 2.0f, match > 85%): 4-Methyl-1-pentaol; 2-ethylpentane; (1S)-2,6,6-trimethyl-bicyclo[3.1.1]hept-2-ene; 2,2-dimethyl-3-methylenenorbornane; (1S)-6,6-dimethyl-2-methylele-bicyclo[3,1,1]heptane; 1,2,4-trimethylbenzene; 1-methyl-4-(1-methylethenyl) cyclohexene; 2-methoxy-quinolin-6(5H)-one; 3,5-dimethyl-loctane; 1-ethyl-4-methylbenzene; 2-propylheptanol; 2-propyldecan-1-ol; and *n*-dodecane, *n*-tridecane, *n*-tetradecane, and 2-phenylphenol. Since these compounds were found at trace levels, they were not included in quantitative determination.

In our study, the average concentrations of benzene and total BTEX measured at non-charcoal-grill sites were 1.82 and 5.79 µg·m⁻³, respectively, whereas those at charcoal grill sites were 2.93 and 10.5 µg·m⁻³, respectively. These values are within the range of those obtained in other studies, for example 1.13 and 6.89 µg·m⁻³ in Michigan, USA [15], and 1.2 and 11.4 µg·m⁻³ in Valencia, Spain [16].

Outdoor concentrations of these compounds are commonly lower than those indoors [15, 16]; yet, at elevated levels, they can markedly deteriorate air quality around restaurants. Significantly, the present study's group B sites were located either amidst residential private houses or near apartment complexes. Thus, there is a significant risk that increased outdoor concentrations of those compounds will result directly or indirectly in adverse health effects for neighboring residents. In fact, placards protesting such businesses often can be seen in these areas.

BTEX and *n*-alkanes are emitted from charcoal itself 5, 6] and from cooking materials [3, 7]. Cooking temperatures higher than 154ºC involve the so-named Maillard reactions, which result in the browning of food and the formation of thermally decomposed or reformed products. Low-temperature cooking, such as stewing in water, is recommended as a means of reducing the formation and emission of hazardous materials.

However, Korean people typically prefer the flavor of fast-cooked charcoal-grilled meat (e.g., beef and pork) over the traditional slow-cooked alternative. Even *bulgogi*, a traditional Korean marinated beef or pork dish, was once cooked in the frying pan but is currently grilled using charcoal. In restaurants, charcoal grilling is carried out mostly by customers themselves at their respective tables. There has been a recent sharp increase in the number of charcoal grill restaurants nationally, making both of the following two risks more dangerous:

1) adverse health outcomes for restaurant customers and workers through exposure to hazardous materials (PAHs, HCAs, VOCs, heavy metals, CO, etc.)
2) secondary adverse health outcomes for pedestrians and residents nearby as the result of contamination of the surrounding outdoor air by emitted pollutants.

In addition to hazardous organic pollutants, charcoal grilling also produces trace hazardous metals such as mercury, chromium, nickel, arsenic, cadmium, lead, selenium, zinc, barium, and others, simply because they are contained in charcoals [17, 18], which, when combusted, release them into the air [19]. These metals have their origins not primarily from nature (material cycles), but mostly from anthropogenic activities (e.g. the recycling of inorganic-preservatives-containing wood, and the addition of metal ingredients to charcoal in the manufacturing process).
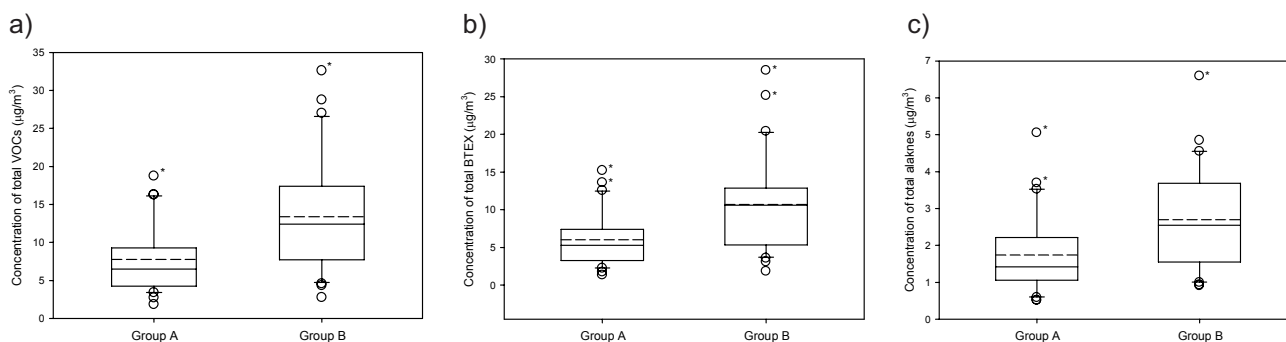

Fig. 3. Box plots comparing concentrations of total VOCs (a), total BTEX (b), and total *n*-alkanes (c) between group A (no charcoal grilling nearby) and group B (at least three charcoal grill restaurants nearby). The dashed and solid lines in each box indicate the mean and median values, respectively. The whiskers (error bars) below and above each box indicate the 10th and 90th percentiles, respectively. The asterisk (*) – marked open circles indicate mild outliers.

Table 2. Matrices of correlation coefficients (*r*) between 10 compounds studied at sites included in groups A and B.

Group A									
VOC***	B	T	EB	*m,p*-X	*o*-X	H	O	N	D	U
B	1.000									
T	0.146	1.000								
EB	0.455	0.559	1.000							
m,p-X	0.279	0.437	0.650*	1.000						
o-X	0.315	0.472	0.881**	0.768**	1.000					
H	0.504	0.524	0.372	0.510	0.502	1.000				
O	0.462	0.583	0.714**	0.553	0.671**	0.672**	1.000			
N	0.462	0.387	0.810**	0.611*	0.726**	0.362	0.640*	1.000		
D	0.230	0.452	0.482	0.694**	0.418	0.440	0.589	0.694**	1.000	
U	0.045	0.353	0.563	0.573	0.615*	0.414	0.617*	0.633*	0.547	1

Group B									
VOC	B	T	EB	*m,p*-X	*o*-X	H	O	N	D	U
B	1.000									
T	0.415	1.000								
EB	0.207	0.561	1.000							
m,p-X	0.032	0.435	0.635*	1.000						
o-X	-0.024	0.477	0.754**	0.819**	1.000					
H	0.376	0.463	0.316	0.612*	0.476	1.000				
O	0.471	0.516	0.334	0.596	0.497	0.636*	1.000			
N	0.295	0.593*	0.418	0.522	0.664**	0.337	0.615*	1.000		
D	0.079	0.447	0.478	0.751**	0.731**	0.508	0.736**	0.812**	1.000	
U	0.241	0.428	0.292	0.497	0.493	0.223	0.594*	0.812**	0.731**	1.000

*The correlation is significant at $\alpha = 0.05$ (2-tailed).
**The correlation is significant at $\alpha = 0.01$ (2-tailed).
***B – benzene, T – toluene, EB – ethylbenzene, X – xylene, H – *n*-hexane, O – *n*-octane, N – *n*-nonane, D – *n*-decane, U – *n*-undecane.

Nonetheless, such cooking activities are not currently regulated and controlled in most countries despite their probable threats to human health, especially via inhalation of polluted indoor air and ingestion of meat containing suspected carcinogens [9]. A duct-ventilation system usually is installed at each table in order to exhaust smoke to the outside air, but the efficiency of this type of setup is not adequate to completely eliminate all indoor air pollutants. Indeed, these ventilation systems typically are not equipped with the types of pollution-reduction devices that effectively capture gas- or particulate-phase pollutants.

The different correlation patterns found for the two groups suggest different emission patterns that result in varying compositions of VOCs. Although it was not easy to determine the characteristic correlation pattern in each group, it is likely that their VOC-formation characteristics vary with the food cooked, the temperature applied, and other factors.

Conclusions

From this study, it was found that outdoor air in areas near charcoal grill restaurants was more contaminated with VOCs such as BTEX and *n*-alkanes than that in areas far from such restaurants. Consequently, pedestrians and residents living near those places can be exposed to elevated levels of pollutants and their secondarily generated by-products. These findings suggest that regulatory action needs to be taken to control the release of air pollutants from charcoal grill restaurants and thereby mitigate urban air pollution.

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