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An investigation on hazardous and odorous pollutant emission during cooking activities

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ABSTRACT

In this study, the emission characteristics of various pollutants (e.g., reduced sulfur compounds (RSCs), aldehydes, volatile organic compounds (VOCs), and organic acids) were investigated in relation to 3 food types (including cabbage, clam, and coffee seeds) and 2 cooking methods (between mild and harsh treatments). The results indicated the strongest emissions from the roasted coffee seeds out of all 6 sample types. Among the pollutant types, the maximum emissions generally came from RSCs followed by aldehydes and acids. Among VOCs, toluene and methyl ethyl ketone were emitted most prominently. As most of these pollutants also represent key odorants, their concentrations are compared through a conversion into odor intensity (OI); the results showed the RSC group as the key odorants along with aldehydes and organic acid compounds. If the sum of odor intensity (SOI) is derived for each sample, they were in the descending order: roasting coffee seeds (6.50), frying cabbage (4.52), brewing coffee (4.14), grilling clam (3.91), boiling clam (3.89), and steaming cabbage (3.21). Their concentration data were also evaluated against regulation guidelines for indoor air quality (IAQ). Comparison of these pollutant data confirms that some cooking approaches can contribute significantly to the build up of nuisance and hazardous pollution concurrently.

1. Introduction

Food cooking is a known source of air pollution and/or odor emissions, as it can release gases and tiny solid particles as by-products [1]. Various pollutants such as volatile organic compounds (VOCs), aldehyde, and H_2S are common components of certain cooking activities [2]. Cooking fumes can contain a list of hazardous pollutants due to incomplete combustion of carbonaceous components in the food material [3,4]. Shields et al. [5] measured different pollutants emitted from cooking foods in relation to various appliances such as ovens, broilers, and griddles. They found the highest levels of emissions from fatty foods cooked at high heat, especially over open flames. Moreover, grilled food items, prepared at extremely high heat, became one of the main causes of air pollution, posing threats to human health [6].

Pollutant emissions from food mainly result from heating and cooking operation through which organic materials in the food are volatilized: under such circumstances, odor and VOC are usually the main concern [2]. However, apart from the odor nuisance, cooking fumes may comprise a wide range of chemical constituents such as oil, fats, aliphatic hydrocarbons, poly-aromatic hydrocarbons, aromatic amines, aldehydes, and elemental carbon [7]. The nature and quantities of pollutants emitted from those sources would highly depend on the cooking stuff, cooking styles, and even on cooking fuel. With the growing awareness of health hazards associated with cooking, the emissions of cooking fume and odor especially from commercial restaurants or household facilities have occasionally become the target of complaints. The problems become much intense and prominent in the urbanized regions, as large-scale restaurants are often placed remarkably close to dwellings.

The objectives of this preliminary study were to quantify the emission of various pollutants released from different food types in combination with different cooking styles. As a primary means to learn more about hazardous pollution from the cooking process, a list of offensive odorants has been selected. This choice of target compounds was basically made to cover a list of major offensive odorants (e.g. reduced sulfur compounds (RSCs), aldehydes, volatile organic compounds (VOCs), organic acids, etc.) regulated by the Korean Ministry of Environment [8]. The results of our study are evaluated further against some of the most well-established regulation guidelines that are recommended to control indoor air quality (IAQ) in relation to the major hazardous pollutants.

2. Materials and methods

The primary target of this study was selected to cover a total of 19 out of 22 compounds that are designated as offensive odorants by the malodor prevention law in Korea [8]. These 19 odorants can

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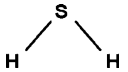
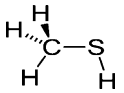
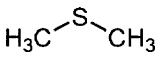
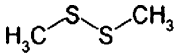
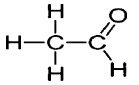
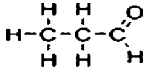
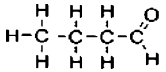
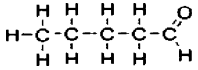
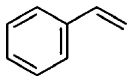
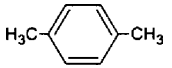
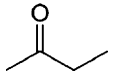
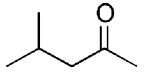
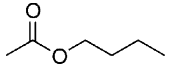
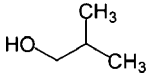
Table 1
The basic physicochemical properties of the target odorants investigated in this study.

Group	Full name	Short name	Chemical formula	Chemical structure	CAS number	Molecular weight ($g\,mol^{-1}$)	Odor threshold[a] (ppb)	Permissible concentration[b] (ppb)
Reduced sulfur compound (RSC)	Hydrogen sulfide	H_2S	H_2S		7783-06-4	34.1	0.41	20
	Methyl mercaptan	CH_3SH	CH_3SH		74-93-1	48.1	0.07	2
	Dimethyl sulfide	DMS	$(CH_3)_2S$		75-18-3	62.1	3.00	10
	Dimethyl disulfide	DMDS	$(CH_3)_2S_2$		624-92-0	94.2	2.20	9
Aldehyde	Acetaldehyde	AA	CH_3CHO		75-07-0	44.0	1.50	50
	Propionaldehyde	PA	C_3H_6O		123-38-6	58.1	1.00	50
	Butyraldehyde	BA	C_4H_8O		123-72-8	72.1	0.67	29
	Isovaleraldehyde	IA	$C_5H_{10}O$		590-86-3	86.1	0.10	3
VOC (aromatic)	Toluene	T	C_7H_8		108-88-3	92.1	330	10,000
	Styrene	S	C_8H_8		100-42-5	104	35	400
	para-Xylene	p-X	C_8H_{10}		106-42-3	106	58	1000
VOC (others)	Methyl ethyl ketone	MEK	C_4H_8O		78-93-3	72.1	440	13,000
	Methyl isobutyl ketone	MIBK	$C_6H_{12}O$		108-10-1	100	170	1000
	Butyl acetate	BuAc	$C_6H_{12}O_2$		123-86-4	116	16	1000
	Isobutyl alcohol	i-BuAl	$C_4H_{10}O$		78-83-1	74.1	11	900

Acid							
Propionic acid	PPA	$C_3H_6O_2$		79-09-4	74.1	5.70	30
Butyric acid	BTA	$C_4H_8O_2$		107-92-6	88.1	0.190	1
Isovaleric acid	IVA	$C_5H_{10}O_2$		503-74-2	102.0	0.078	1
Valeric acid	VRA	$C_5H_{10}O_2$		109-52-4	102.0	0.037	0.9

[a] Source of threshold values: Nagata (2003) [23]. Odor thresholds measured by the triangle odor bag method.
[b] According to malodor prevention law in Korea (KMOE, 2008 [8]).

be divided into 4 chemical groups: (1) RSCs (H_2S, CH_3SH, DMS, and DMDS), (2) aldehyde (acetaldehyde (AA), propionaldehyde (PA), butyraldehyde (BA), and isovaleraldehyde (IA)), (3) VOCs (toluene (T), styrene (S), para-xylene (p-X), methyl ethyl ketone (MEK), methyl isobutyl ketone (MIBK), butyl acetate (BuAc), and isobutyl alcohol (i-BuAl)), and (4) volatile fatty acids (propionic acid (PAd), butyric acid (BAd), isovaleric acid (IAd), and valeric acid (VAd)). Among three of the original 22 listed offensive odorants, ammonia and trimethylamine were not analyzed in this study, while valeraldehyde was not detected in any of the samples. The basic physicochemical properties (e.g., chemical formula, chemical structure, molecular weight, CAS number, etc.) of all target compounds are summarized briefly in Table 1.

2.1. Sample collection

In order to measure odorants released from cooking activities, we mainly focused on three food types including vegetables (cabbage), sea food (clam), and seeds (coffee seeds). These foods are selected for this study, mainly because they are well known for producing unique odors of their own during fire-based cooking. In the course of this study, 100–200 g of three food materials were taken for this investigation by applying 2 types of cooking approaches between mild (steaming, boiling, and brewing) and harsh treatments (oil-based frying, grilling, and roasting) (Table 2). To simplify comparison of the measurement data, we assigned them with two letter acronyms for each food (cabbage (CA), calm (CL), and coffee (CO)) and numbers of 1 (mild) and 2 (harsh) for treatment type (Table 2). As a result, a total of 6 different samples were collected such as: (1) CA-1 and CA-2, (2) CL-1 and CL-2 and (3) CO-1 and CO-2 (Fig. 1). All treatment type 1 samples were collected inside the laboratory, all cooking activities for type 2 samples were carried out in the open space on the rooftop of our lab building. The stripped gas samples released from each cooking process were collected into 10 L Tedlar bags with the aid of lung sampler (ACEN Co. Ltd., Korea) for the analysis of the target compounds. The wind flow during the sampling day was fairly low (below 1 m s^{-1}), and the collection of type 2 samples was made inside paper board cover to minimize the dilution effect. In addition, the influence of wind speed or air exchange rates was not considered for the sample collection made in the laboratory. The cooking for all three food types was done at medium flame on a liquefied petroleum gas (LPG) stove without using any oil or condiments.

2.2. RSCs analysis

The analysis of RSCs was done by gas chromatography (GC) equipped with a pulsed flame photometric detector (PFPD) interfaced with a multi-function thermal desorber (TD) system with an air server (AS) unit. Details of the operating conditions for the RSC analysis have been listed in Table 3(a). The analytical procedures for RSCs in ambient air samples have been described in a number of our previous publications [9,10]. The detection limits (DL) of the system fell in the range of 0.5 (or 0.12 ppb (DMDS))–0.7 pg (or 0.52 ppb (H_2S)) (in a sampling volume of 120 mL). If the precision of this method is evaluated in terms of relative standard error (RSE), it generally ranges from 1.35 (H_2S) to 4.25% (DMDS).

2.3. VOC analysis

The combination of GC with mass spectrometry (MS) system coupled with a multi-function TD was used for the analysis of VOC odorants. The samples were extracted by TD to the system from the Tedlar bag. The TD device comprises a desorption oven connected to a Peltier-cooled sorbent packed cold-trapping system. Chromatographic separation was achieved by Vocol column (60 m × 0.32 mm i.d. and 1.8-μm film thickness: Supelco) at a column flow rate of

Table 2
The basic information of 6 sample types investigated in this study and their assortment by the combination of (2) cooking styles and (3) food materials.

Sample ID	Cooking style	Name	Cooking method	Weight (g)	Place of origin	Sample collection site
CA-1	Mild (steaming)	Cabbage	Steam	200	Korea	
CL-1		Clam	Boil	100	Korea	Laboratory
CO-1		Coffee	Brew	200	Brazil	
CA-2	Harsh (frying)	Cabbage	Fry	200	Korea	
CL-2		Clam	Grill	100	Korea	Outside
CO-2		Coffee	Roast	200	Brazil	

1.2 mL min^{-1} (99.9% pure He as carrier gas). Detailed operating conditions of this system are listed in Table 3(b). The DL values of the VOCs fell in the range of 1.27 (0.31 ppb (MIBK))–1.81 ng (0.38 ppb (BuAc)). The precision of the method if expressed in RSE, varied in the range of 3.1% (MEK)–5.2% (BuAc).

2.4. Volatile fatty acid analysis

A TD system interfaced with a GC–flame ionization detector (FID) was used for the analysis of volatile fatty acids (VFA) (refer to Table 3(c)). The collection of all the acid components was initially made via Carbopack X tube (60/80 mesh, Supelco, PA, USA) samples at a flow rate 200 mL min^{-1} for 5 min with the help of a mini pump (SIBATA, Japan). The analysis of VFA was made in a manner analogous to those of RSCs in that the GC system is interfaced with TD. The DL values of the acid compounds were 0.82 (0.39 ppb PPA), 0.60 (0.20 ppb (BTA)), 0.50 (0.14 ppb (IVA)), and 0.60 ng (0.21 ppb (VRA)). The precision of VFA by the TD based analysis was computed in the range of 4.3–6.8%.

Fig. 1. A list of photographs showing the sample collection procedures of 2 different cooking styles for 3 different food materials.

Table 3
The operational conditions of all instrumental systems employed in this study.

(a) GC/TD system for RSC analysis					
[1] GC (DS 6200, Donam Instrument, Korea) system					
(i) Oven			(ii) Detector (PFPD: Model 5380, O.I. Analytical, USA)		
Initial temp:	80	°C	Detector temp.:	250	°C
Ramp:	20	°C min^{-1}	Air(1)/air(2): flow:	10	mL min^{-1}
Final temp:	200	°C	H_2 flow:	11.5	mL min^{-1}
Initial hold:	4.5	min			
Final hold:	9.5	min			
Total time:	20	min	(iii) Column (BP-1, SGE, Australia)		
			Film thickness:	5	μm
			Length:	60	m
			Diameter:	0.32	mm
[2] Thermal desorber (UNITY, Markes International, Ltd., UK)					
Cold trap:	Carbopack B + Silica Gel = 1.5: 2.5 (volume ratio)				
Split ratio:	10:01		Trap low temp.:	−15	°C
Split flow:	15	mL min^{-1}	Trap high temp.:	250	°C
Hold time:	5	min	Flow path temp:	80	°C
(b) GC/MS system for VOC analysis					
[1]. GC/MS (SHIMADZU GCMS-QP2010, Japan)					
(i) Oven			(ii) Detector (MS)		
Initial temp:	35	°C	Ionization mode:	EI (70 eV)	
Hold time:	4	min	Ion source temp.:	200	°C
Ramping rate:	4	°C min^{-1}	TIC scan range:	35∼250	*m*/*z*
Final temp:	200	°C	Threshold:	100	
Hold time:	10	min			
Carrier gas:	He	99.90%			
			(iii) Column (Vocol, PA, USA)		
			Diameter:	0.32	mm
			Length:	60	m
			Film thickness:	1.8	μm
[2] Thermal desorber (UNITY, Markers International Ltd., UK)					
Cold trap:	Carbopack B+ Tenax				
Split ratio:	20		Trap low:	5	°C
Split flow:	5.0	mL min^{-1}	Trap high:	300	°C
Hold time:	5.0	min	Flow path temperature	120.0	°C
(c) GC/FID system for organic fatty acid analysis					
[1] GC (Varian 450-GC, USA)					
(i) Oven			(ii) Detector (FID)		
Initial temp:	50	°C	Detector temp:	240	°C
Ramping rate:	6	°C min^{-1}	H_2/air flow:	30	mL min^{-1}
Final temp:	230	°C	N_2 flow:	29	mL min^{-1}
Initial and final hold:	5	min			
			(iii) Column (CP-WAX, J&W, CA, USA)		
			Film thickness:	1.8	μm
			Length:	60	m
			Diameter:	0.25	mm
[2] Thermal desorber (UNITY, Markes International Ltd., UK)					
Cold trap:	Carbopack X tube (60/80 mesh)		Trap low temp.:	5	°C
Desorption temp.:	300	°C	Trap high temp.:	300	°C
Hold time:	10	min	Flow path temp:	120	°C
Cold trap hold time:	5	min	Split ratio:	10:1	
Valve temp:	120	°C	carrier gas flow:	1.5	mL min^{-1}
(d) HPLC (Series 1500, Lab Alliance, USA)/UV system for carbonyl compounds analysis					
(i) Injector			(iii) UV detector (Model 500, Lab Alliance, USA)		
Volume:	20	μL	Wavelength:	360	nm
(ii) Pump			(iv) Column (C_{18}, Hichrom, UK)		
Flow rate:	1.5	mL min^{-1}	Column dimensions:	250 × 4.6	mm
Mobile phase:	Acetronitrile:water	70:30	Particle size:	5	μm
Analysis time:	15	min	Pore size:	300	Å
			Temp:	20	°C
			Packing type:	Monomeric	

2.5. Carbonyl analysis

The analysis of carbonyl compounds was carried out by high performance liquid chromatography (HPLC) equipped with a UV detector and dsCHROM software (for peak integration). The basic analytical conditions of the HPLC system are described in Table 3(d). Air samples were passed through Lp DNPH cartridges (Supelco, USA) at a normal set-up value of 10 min (at a fixed sampling flow rate, 0.8 L min^{-1}) via a Sep-Pak ozone scrubber (Waters, USA). After that, the cartridges were eluted slowly with 5 mL methanol and filtered through 0.45 μm, 13 mm, GHP Acrodisc filters (PALL, NY, USA) into a 25 mL capacity borosilicate glass volumetric flask. The

eluate was manually injected into the HPLC system equipped with a 20 μL sample loop. The DL values were 19.1 (or 0.71 ppb (AA)), 14.1 (or 0.77 ppb (PA)), and 13.9 (or 0.39 ppb (BA)), and 15.2 ng (or 0.49 ppb (IA)). For the mixing ratios provided in the parenthesis, we assumed a total sampling volume of 15 L. The precision of analysis, if assessed in terms of RSE, tended to vary in the range of 0.51% (AA)–2.16% (IA).

3. Results and discussion

3.1. General pattern of odorous emission

Cooking activities can generate highly unique, and sometimes unpleasant odors arising from the chemical reactions. Odors generated from the cooking processes are usually a mixture of various organic and inorganic compounds at low concentrations [7]. Most of these compounds are reduced carbon and/or sulfur compounds such as aldehyde, ketone, alcohols, acids, sulfides, and hydrogen sulfide which are easily biodegraded [11]. In some cases, the odors may also be caused by VOCs, which are less biodegradable. The objectionable odors from cooking activities are generally a result of the physical processing of foods usually associated with thermal processing steps (such as evaporative condensation, heating, drying, or smoking of foods) [12].

As summarized in Table 4, among the six types of matching pairs between food materials and cooking processes, significant amounts of odorants were released from certain food types and cooking methods. Comparison of the data between 6 samples

Table 4
Summary of odorant levels released from diverse cooking activities.

(a) Concentration of odorous compounds determined from replicate analysis ($n = 2$) of individual samples collected during cooking process (ppb).

Compound	Matching pair of food material and cooking type[a]					
	CA-1	CL-1	CO-1	CA-2	CL-2	CO-2
H_2S	0.86	0.20[b]	0.20	0.20	39.6	2398
CH_3SH	0.15	0.15	13.5	63.8	0.15	2070
DMS	9.44	0.26	16.9	25.6	31.3	98.7
DMDS	1.20	0.06	4.32	9.34	35.5	24.5
AA	12.0	18.7	153	12.5	253	5233
PA	0.39	2.81	31.8	5.40	8.65	366
BA	0.39	0.39	77.6	15.3	12.9	458
IA	0.44	0.44	0.44	0.44	0.44	600
S	0.37	0.31	0.36	0.07	0.20	8.36
T	26.3	19.8	24.0	51.2	51.1	123
p-X	1.62	1.51	1.95	1.57	1.99	0.03
MEK	3.21	5.45	52.6	3.21	28.2	964
MIBK	0.04	0.48	0.04	0.04	0.04	0.04
BuAc	0.44	0.04	0.04	0.04	0.04	0.04
i-BuAl	0.09	0.09	3.08	0.09	3.91	0.09
PPA	2.27	2.50	5.84	4.39	36.1	695
BTA	0.06	0.20	0.06	0.06	5.11	67.0
IVA	3.46	5.75	15.9	0.05	1.97	132
VRA	0.06	0.06	0.06	0.14	0.12	8.39

(b) Odor intensity (OI) distribution of the target compounds.

Compound	Function[c]	Odor intensity					
		CA-1	CL-1	CO-1	CA-2	CL-2	CO-2
H_2S	$Y = 0.950 \log X + 4.14$	1.23	–[d]	–	–	2.81	4.50
CH_3SH	$Y = 1.250 \log X + 5.99$	–	–	3.65	4.50	–	6.38
DMS	$Y = 0.784 \log X + 4.06$	2.47	1.25	2.67	2.81	2.88	3.27
DMDS	$Y = 0.985 \log X + 4.51$	1.63	–	2.18	2.51	3.08	2.92
AA	$Y = 1.010 \log X + 3.85$	1.91	2.10	3.03	1.93	3.25	4.58
PA	$Y = 1.010 \log X + 3.86$	–	1.28	2.35	1.57	1.78	3.42
BA	$Y = 1.060 \log X + 4.23$	–	–	3.05	2.31	2.23	3.87
IA	$Y = 1.350 \log X + 6.01$	–	–	–	–	–	5.71
S	$Y = 0.790 \log X + 2.53$	–	–	–	–	–	0.89
T	$Y = 1.380 \log X + 4.60$	2.42	2.25	2.36	2.82	2.82	3.34
p-X	$Y = 1.570 \log X + 2.44$	–	–	–	–	–	–
MEK	$Y = 1.850 \log X + 0.15$	–	–	–	–	–	0.12
MIBK	$Y = 1.650 \log X + 2.27$	–	–	–	–	–	–
BuAc	$Y = 1.140 \log X + 2.34$	–	–	–	–	–	–
i-BuAl	$Y = 0.790 \log X + 2.53$	–	–	0.62	–	0.70	–
PPA	$Y = 1.380 \log X + 4.60$	0.95	1.01	1.52	1.35	2.61	4.38
BTA	$Y = 1.290 \log X + 6.37$	–	1.60	–	–	3.41	4.86
IVA	$Y = 1.090 \log X + 5.65$	2.97	3.21	3.69	–	2.70	4.69
VRA	$Y = 1.580 \log X + 7.29$	–	–	–	1.20	1.10	4.01
SOI		3.21	3.89	4.14	4.52	3.91	6.50

[a] CA-1, steaming cabbage; CL-1, boiling clam; and CO-1, brewing coffee seeds; CA-2, frying cabbage; CL-2, grilling clam; CO-2, roasting coffee seeds.
[b] Underlined numbers denote the concentration data equivalent to detection limit (DL).
[c] Nagata [23], odor intensity (Y) and odorant concentration (ppm) (X).
[d] No numeric values are shown for the cases with Negative OI values.

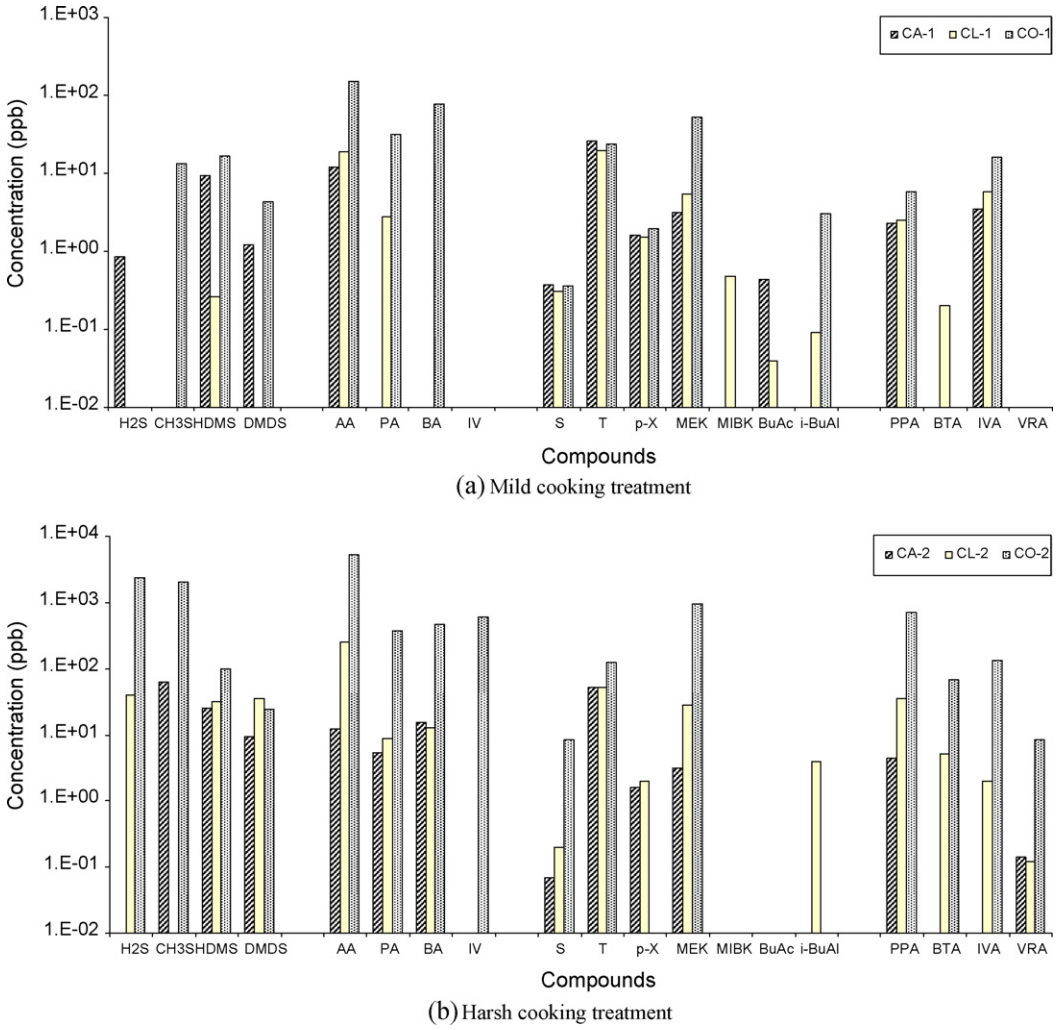


Fig. 2. Comparison of the odorant concentration levels (ppb) released from food samples between mild and harsh cooking treatments.

indicates that odorant emissions were predominated by roasting coffee seeds. Roasting of coffee seeds can induce Maillard reaction [13]. The high temperature and elevated pressure inside the seed are known to trigger a vast number of chemical reactions that can alter or create volatile aromatic compounds, acids, and other critical flavor components [13]. However, considerable amount of odorants was also released from (1) frying cabbage: 63.8 ppb (CH_3SH), 25.6 ppb (DMS), and 51.2 ppb (toluene), (2) grilled clam: 253 ppb (acetaldehyde), 39.6 ppb (H_2S), 35.5 ppb (DMDS), 31.3 ppb (DMS), and 36.1 ppb (propionic acid), and (3) brewing coffee: 31.8 ppb (propoinaldehyde), 77.6 ppb (butyraldehyde), 52.6 ppb (MEK), and 15.9 ppb (isovaleric acid) (Table 4(a)). Mikuła [14] reported higher levels of sulfur in cabbage leaves relative to other vegetables in Poland. In this study, great amount of toluene emission observed from frying cabbage can be ascribed to those taken up from the atmosphere by plant leaves and other aboveground parts [15]. Gorna-Binkul et al. [16] also carried out a survey of toluene using shop-bought fruits and vegetables in Poland and found cabbage leaves to contain high levels of toluene (e.g., 228 ppb). Wilmot and Vetter [17] concluded that sulfide oxidation occurs in the animal tissue of clam (instead of the symbiotic bacteria). This might be the reason to observe moderate emissions of RSCs from grilling clams. In another study conducted in Korea, it was also found that clams contained significantly high amount of odorous compound like propionic acid [18].

RSCs were the most abundant form in CO-2 followed by CL-2 and CA-2 samples. It is interesting to note that isoveleraldehyde was observed only from CO-2 among all 6 samples. Likewise, acetaldehyde was recorded only from CA-1. Among the VOCs, toluene was the most dominant compound followed by MEK. As expected, the highest concentrations of toluene and MEK were found in CO-2 as 123 and 964 ppb, respectively (Fig. 2). The emission concentrations of styrene and para-xylene were not significantly large from most samples. Nevertheless, the emission concentrations for MIBK, butyl acetate, and isobutyl alcohol were typically seen below the DL values from most samples. In case of fatty acid compounds, their highest discharges were also found from CO-2. Notably large quantities of acid compounds were also detected from the CL-2 sample. Among the acid compounds, propionic and isovaleric acids were released from almost all the samples, while it was not the case for butyric and valeric acids.

3.2. Evaluation of data in terms of odor intensity

Odor nuisance is generally defined by the four factors: frequency, intensity, duration, and offensiveness. These key properties can be defined briefly as follows. Frequency refers to the number of times an odor occurs, intensity refers to the strength of an odor, duration refers to the period of time an odor is encountered, and offensiveness refers to the character or hedonic tone of the odor (pleasant or unpleasant) [19,20]. A quantitative description of odor

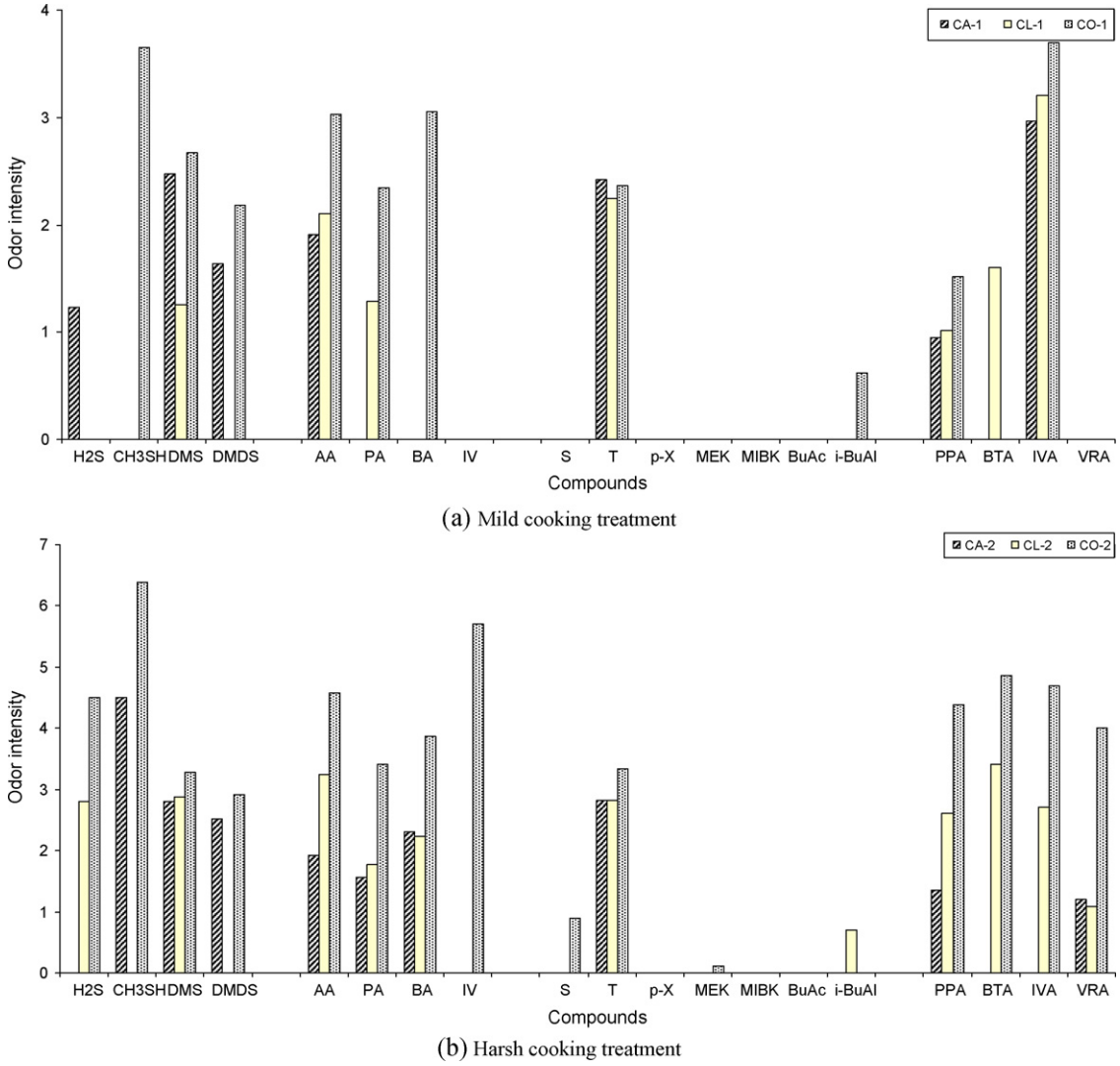


Fig. 3. Comparison of the odor intensity (OI) values between mild and harsh cooking treatments.

exposure is limited by the combined effects of few factors due to the complexity of odorant mixing and/or the delicacy of its detection by the human nose [21]. In this respect, the use of odor intensity (OI) concept is a highly meaningful approach, as it provides a parallel means to evaluate concentration data of the perceived odorants through numerical conversions [22].

For this purpose, the concentration data of each odorant measured in this study were converted into the OI with the varying index numbers with the aid of empirical equations developed by Nagata [23]. The OI scaling of 0 through 6 can be distributed as follows: 0 (no odor), 1 (very weak), 2 (weak), 3 (distinct), 4 (strong), 5 (very strong), and 6 (intolerable) [24]. However, as the OI values of less abundant compounds are occasionally converted into negative range, such values were disregarded for simplicity. Concentrations of all the offensive odorants measured during the entire study period are summarized along with the computed OI values in Table 4(b).

A brief inspection of the data indicates that the magnitude of the data expressed in terms of both concentration and OI terms differs greatly among food types and cooking processes. Based on the OI convention, most of the non-VOC groups (e.g., RSCs, aldehyde, and fatty acids) exhibit positive OI values in various cooking operations, while most VOCs do not (Fig. 3). In case of VOCs, only toluene was detected consistently across all 6 sample types investigated in this study. This result thus suggests that the VOCs are less likely to contribute to the strengths of odor in cooking activities. The largest OI value is observed from CH_3SH (6.38) followed by isovaleradehyde (5.71) from roasting coffee seeds (Fig. 3). The overall evaluation of OI ratings thus confirms that RSCs are the dominant odorant constituents contributing to the nuisance of cooking activities for all 3 food materials.

As can be expected from the concentration data, the patterns of OI ratings confirm the roasting coffee seeds to be dominant among all 6 sample types. In an effort to assess the relative intensity of odorants measured in this study, the concept of odor threshold can also be employed. The threshold odorant concentration (TOC) of a pure compound in air can be defined in a number of ways such as the lowest concentration that can be perceived by the 50% of the tested population [25]. In this study, we adopted TOC values reported by Nagata [26] as the main reference for such comparison (Table 1) [4]. If these TOC criteria are applied to our data, the highest frequency of the measured data exceeding such criteria was observed from most chemical groups, e.g., RSC, aldehyde, and acid compounds (except the VOCs). This implies that the odor nuisance during the sample collection was moderately strong. (Our team also experienced high odor intensity, while collecting the samples.) The frequency of such exceedance cases for the 6 samples was counted as: 4 (CA-1), 4 (CL-1), and 8 (CO-1), 7 (CA-2),

Table 5
Results of correlation analysis between target compounds considered in this study.

(a) Results derived using all samples[a].

		H_2S	CH_3SH	DMS	DMDS	AA	PA	BA	S	T	X	MEK	PPA	BTA	IVA
	r[b]	1													
	p[c]														
H_2S	*N*[d]	3													
	r	–[e]	1												
	P														
CH_3SH	*N*	2	3												
	r	0.975	0.997*[f]	1											
	P	0.142	0.048												
DMS	*N*	3	3	6											
	r	0.217	0.976	0.541	1										
	P	0.861	0.139	0.346											
DMDS	*N*	3	3	5	5										
	r	0.999*	0.999*	0.957**[g]	0.398	1									
	P	0.017	0.029	0.003	0.507										
AA	*N*	3	3	6	5	6									
	r	–	0.996	0.949*	0.249	0.998**	1								
	P		0.055	0.013	0.750	1.3E−04									
PA	*N*	2	3	5	4	5	5								
	r	–	0.989	0.957*	0.194	0.989**	0.997**	1							
	P		0.097	0.042	0.806	0.010	0.003								
BA	*N*	1	3	4	4	4	4	4							
	r	0.999*	0.999*	0.942**	0.352	0.998**	0.998**	0.993**	1						
	P	0.021	0.033	0.005	0.560	4.3E−06	9.3E−05	0.007							
S	*N*	3	3	6	5	6	5	4	6						
	r	0.972	0.969	0.988**	0.563	0.939**	0.923*	0.902	0.923**	1					
	P	0.149	0.157	0.001	0.323	0.005	0.025	0.098	0.009						
T	*N*	3	3	6	5	6	5	4	6	6					
	r	–	–	0.613	0.578	0.946*	0.656	0.394	0.120	0.251	1				
	P			0.272	0.422	0.015	0.344	0.742	0.847	0.684					
X	*N*	2	2	5	4	5	4	3	5	5	5				
	r	1.0**	0.998*	0.953**	0.372	0.999**	0.999**	0.994**	0.998**	0.931**	0.870	1			
	P	0.006	0.042	0.003	0.537	5.7E−07	1.5E−05	0.006	2.6E−06	0.007	0.055				
MEK	*N*	3	3	6	5	6	5	4	6	6	5	6			
	r	0.992	0.999*	0.958**	0.407	0.999**	0.995**	0.986*	0.997**	0.943**	0.709	0.998**	1		
	P	0.080	0.015	0.003	0.496	1.1E−07	3.0E−04	0.014	6.8E−06	0.005	0.180	3.8E−06			
PPA	*N*	3	3	6	5	6	5	4	6	6	5	6	6		
	r	–	–	0.969	–	0.999*	0.650*	–	0.996*	0.973	–	0.998*	0.999*	1	
	P			0.158		0.017	0.033		0.049	0.149		0.029	0.015		
BTA	*N*	2	2	3	2	3	3	2	3	3	2	3	3	3	
	r	0.999*	–	0.949*	0.280	0.994**	0.999**	0.999*	0.996**	0.940*	0.300	0.997**	0.992**	0.996	1
	P	0.015		0.013	0.719	4.8E−04	0.001	0.024	2.9E−04	0.017	0.700	1.4E−04	0.001	0.058	
IVA	*N*	3	2	5	4	5	4	3	5	5	4	5	5	3	5
	r	–	–	0.987*	0.089	0.999*	0.999**	0.999**	0.999**	0.999**	–	0.999*	0.999*	–	–
	P			0.04	0.943	0.027	0.006	0.002	0.010	0.001		0.016	0.027		
VRA	*N*	2	2	3	3	3	3	3	3	3	2	3	3	2	2

(b) Summary of correlation analysis for each sample group.

Sample source	Frequency of matching pairs at 2 significance levels		The total number of possible matching pairs
	0.01	0.05	
All	33	25	105
Sample type 1 (mild)	2	2	28
Sample type 2 (harsh)	11	25	45

[a] IA, MIBK, BuAc, and i-BuAl are not considered for the correlation analysis as most of the values are below detection limit.
[b] Pearson's correlation coefficient.
[c] Probability (2 tails significance).
[d] No of data.
[e] Not computed when *N* = 1 and 2.
[f] *Correlation is significant at the 0.05 level (2-tailed).
[g] **Correlation is significant at the 0.01 level (2-tailed).

10 (CL-2), and 13 (CO-2) out of all possible 19 cases (or pollutants measured).

As a simple approach to briefly assess the overall contribution of individual components released from a given sample to odor formation, the OI values of each individual compound were bound together to derive the total odor strength for each sample type in terms of "sum of odor intensity (SOI)". For the derivation of SOI term, the following equations were employed in this study [27]:

$$\mathrm{SOI} = \log\left(\sum 10^{\mathrm{OI}(i\mathrm{th})}\right) = \log\left(10^{\mathrm{OI}(i\mathrm{th})1} + 10^{\mathrm{OI}(i\mathrm{th})2} + 10^{\mathrm{OI}(i\mathrm{th})3} + \ldots + 10^{\mathrm{OI}(i\mathrm{th})n}\right), \quad \text{where} \quad \mathrm{OI}(i\mathrm{th}) = \log 10^{\mathrm{OI}(i\mathrm{th})}$$

In terms of the SOI, the strength of odorant emission tends to peak from roasting coffee seeds (6.50) followed by frying cabbage (4.52), brewing coffee (4.14), grilling clam (3.91), boiling clam (3.89), and steaming cabbage (3.21). In all cases, the odor strengths of frying (harsh) style appeared to be stronger than boiling (mild) style without any single exception. In one of the previous studies, both food and oil type as well as temperature were seen to exert a significant effect on cooking emission patterns [1].

3.3. Factors affecting odorant emission from cooking activities

In order to learn more about the factors governing the odorant emissions from various foods and cooking processes, Pearson's correlation analysis was done using all concentration data (Table 5(a)). According to this analysis, 58 out of 105 matching pairs were correlated significantly ($P < 0.05$). If we divide the results by two simple criteria of P values less than 0.05 and 0.01, 25 and 33 cases fell into such category, respectively. Isovaleraldehyde, MIBK, butyraldehyde, and isobutyl alcohol are not considered for the correlation analysis, as most of them were measured below the detection limit. It is interesting to note that H_2S did not show strong correlations with the other sulfur compounds, while displaying significant correlations with many other odorants (e.g., acetaldehyde, styrene, MEK, and isovaleric acid). The observed pattern of H_2S appears to be unusual in that most RSCs generally show strong interactions with each other. For instance, Wu and his colleagues measured volatile organic sulfur compounds from food wastes during laboratory-controlled aerobic decomposition and found significant correlations between them [28]. In contrast, most aldehyde compounds showed good correlations among themselves. Among the VOCs, styrene, toluene, and MEK were also seen to be strongly correlated with each other. However, para-xylene showed correlation only with acetaldehyde. In case of acids, only propionic acid showed significant correlations with butyric, isovaleric, and valeric acids. Except for some unique patterns described above, most target compounds frequently showed significant correlations with each other such as: H_2S (4), CH_3SH (5), DMS (10), acetaldehyde (13), propionaldehyde (10), butyraldehyde (8), styrene (12), toluene (8), MEK (12), propionic acid (11), butyric acid (5), isoveleric acid (9), and valeric acid (8).

If the results are compared between different sample types, fairly strong correlations are observed for most matching pairs in frying methods relative to steaming cooking style (Table 5(b)). In order to investigate the air pollutants emission from different cooking styles, Lee et al. [29] investigated indoor air quality of four restaurants (a Korean barbecue style restaurant, a Chinese hot pot restaurant, a Chinese dim sum restaurant, and a Western canteen) in Hong Kong. They were able to confirm significant correlations between many VOCs.

3.4. Health hazard of cooking pollutants

Historically, people have been cooking foods for more than 100,000 years. Until recently scientists did not carefully probe the contents of cooking smoke. It is however time to wonder whether and how that smoke might contribute to air pollution or pose health hazards. As most foods do not contain large quantities of toxins, there have been only limited pieces of information concerning the proven health risks associated with cooking fumes. It has been assumed that the tiny particles released in the form of cooking smoke could be lodged deep into the lungs, where they might cause cancer or other problems [30]. Several studies have implicated that domestic exposure to cooking fumes as a possible risk factor, although the exact identification and quantities of carcinogens have yet to be identified [31,32]. One may note the fact that the rate of lung cancer in Chinese women was high relative to other countries [29]. It is suspected that the high-temperature wok cooking with unrefined Chinese rapeseed oil may be one of the responsible factors for that. The volatiles emitted from unrefined cooking oils were reported to be mutagenic [5].

Evaluation of sensory and health effects from indoor air exposure is hampered by the limited number of specific indoor air standards and guidelines with respect to cooking. The evaluations are made mainly based on three categories [33]. One category includes annoyance due to odor. Another category includes the irritation effects on the eyes and the upper respiratory tract, termed sensory irritation [34,35]. Finally, the genotoxically carcinogenic substances comprise the third category [36]. The potential health effects of organic chemicals are influenced by many factors including the duration of the exposure, time of day, day of week, intensity, and frequency of exposure [37].

In Table 1, the maximum allowable emission concentrations of all target compounds investigated in this study were provided by referring to the malodor prevention law in Korea [8]. If our data are examined in relation to these criteria, the concentrations of the major odorants were much higher than such criteria in some samples, especially roasted coffee seeds. Moreover, if the number of cases exceeding this malodor prevention guideline is evaluated across each compound for all 6 samples, the results can be summarized as follows: 2 (H_2S), 3 (CH3SH), 4 (DMS), 3 (DMDS), 3 (AA), 1 (PA), 2 (BA), 1 (IV), 2 (PPA), 2 (BTA), 5(IVA), and 1 (VRA). It is also worth mentioning that

Table 6
Indoor air guideline values (in ppm) for some target compounds investigated in this study.

Compound name	ACGIH[a]	OEHHA[b]	WHO[c]	MHLW[d]	HK[e]	ATSDR[f]	Maximum emission from the samples
AA	25	0.005	0.278	0.267	–	–	5.23
T	50	0.079	0.069	0.069	0.289	3.00	0.123
S	20	0.212	0.061	0.052	–	0.06	0.008
p-X	100	0.162	1.11	0.201	0.334	0.23	0.002

[a] Threshold limit value set by American Conference of Governmental Industrial Hygienists (ACGIH), 2004 [38].
[b] Non-cancer chronic reference exposure level, Office of Environment Health Hazard Assessment (OEHHA), 2007 [39].
[c] World Health Organization (WHO) Guidelines for Air Quality, 1999 [40].
[d] IAQ Guidelines by Ministry of Health, Labor, and Welfare (MHLW) of Japan, 2004 [41].
[e] Guidelines for Good Class IAQ set by the Government of the Hong Kong Special Administrative Region, 2003 [42].
[f] Agency of Toxic Substance and Disease Registry (ATSDR), USEPA, 1998 [43].

none of the VOCs were recorded to exceed this type of guideline.

The emission concentrations of the target compounds can affect occupant's comfort and health. In order to design acceptable indoor environments, practitioners refer to standards and guidelines developed by a variety of agencies. Table 6 summarizes some of the most well-established regulation guidelines that are recommended to control indoor air quality (IAQ) in relation to the major hazardous pollutants. Here note that the emission concentrations for minor compounds are not listed as a potential health risk. In several occasions, the emission concentrations of acetaldehyde and toluene from these samples exceeded the guideline values set by non-cancer chronic reference exposure level by Office of Environment Health Hazard Assessment (OEHHA), World Health Organization (WHO) guidelines for air quality, and indoor air quality Guidelines by Ministry of Health, Labor, and Welfare (MHLW) of Japan. Moreover, human exposure to the target compounds can cause irritation of the eyes and respiratory system, mood swings, headaches, nausea, and drowsiness [44]. Nevertheless, Yu et al. [45] informed that cumulative exposure to cooking (by means of any form of frying) could increase the risk of lung cancer of the nonsmoking women.

4. Conclusion

A number of air pollutants are released during cooking activities. Some of them can cause odor nuisance, while being hazardous to human health. Among the six types of matching pairs between food materials and cooking processes investigated in this study, odorant emissions prevailed by roasting coffee seeds followed by brewing coffee, frying cabbage, and grilled clam. The concentrations of the pollutants released from roasting coffee seeds were significantly high relative to other sample types obtained during the cooking periods. The cooking gases produced from food materials considered in this study were generally characterized by higher abundances of RSCs and aldehydes in contrast with most VOCs.

The magnitude of odor intensity from these cooking samples varied rather dynamically between food types and between cooking processes. The highest OI of the target compounds was found most frequently from roasted coffee seeds. RSCs were the prominent odorants responsible for the odor nuisance along with aldehydes and acid compounds from most cooking activities investigated in this study. In case of VOCs, only toluene showed positive OI values for all 6 samples. If the sum of odor intensity (SOI) values are derived for all 6 sample types investigated in this study, they can be listed as follows: roasting coffee seeds (6.50), frying cabbage (4.52), brewing coffee (4.14), grilling clam (3.91), boiling clam (3.89), and steaming cabbage (3.21). Moreover, most target compounds showed significant correlations with each other in Pearson's correlation analysis. In few cases acetaldehyde and toluene exceeded the guideline values set for health concern (e.g., by OEHHA and WHO). The results of our study suggest that foods produced by different cooking methods can cause odor nuisance and pose health threats to a varying degree.

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