



Release of offensive odorants from the combustion of barbecue charcoals

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ABSTRACT

A number of offensive odorants including volatile organic compounds (VOCs), reduced sulfur compounds (RSCs), carbonyls, and ammonia were measured along with several reference pollutants (like benzene (B), CS₂, SO₂, CO, and total hydrocarbon (THC)) from combusted fumes of barbecue charcoals produced from five different countries (Korea, China, Indonesia, Malaysia, and the US). Although the emission concentrations of most odorants were generally below the reference guideline set by the malodor prevention law in Korea, the mean concentration of some aldehydes (acetaldehyde, propionaldehyde, and isovaleraldehyde) and ammonia exceeded those guidelines. As such, aldehydes were the most dominant odorant released from charcoal combustion followed by VOC and ammonia. If odorant levels of charcoal products are compared, there are great distinctions between the products of different countries. If comparison is made using the concept of the sum of odor intensity (SOI), the magnitude of SOI for the charcoal products from the five different countries varied in the order of 4.30 (Korea), 3.10 (Indonesia), 2.97 (China), 2.76 (Malaysia), and 2.76 (the US).

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1. Introduction

Charcoal is a conventional term for a char obtained from wood, peat, coal, or some related natural organic material through carbonization. This process allows for the conversion of organic matter to solid residues with increased elemental carbon content via pyrolysis [1]. As charcoal has high heating values with low smoke and dust relative to wood [2], it is used extensively for cooking purposes, especially in barbecuing. As an integral part of leisure activities and/or ceremonies, charcoal barbecue foods are prepared for different cultural activities in many countries. In some countries (i.e., Korea, China, etc.), traditional barbecue restaurants are also very popular, wherein charcoal is used as the main fuel for barbecuing. Although charcoal is not the prime source of cooking energy in many developed countries, it remains as one of the most commonly selected fuels for cooking in the present world.

Charcoal is made up of various types of organic and inorganic compounds such as hydrocarbons, sulfur, water, and oxygen along with numerous trace elements [1]. Recent investigations of charcoals also revealed that its combustion fume can act as a potential source of volatile organic compounds (VOCs), carbonyls, trace metals (including mercury), and polycyclic aromatic hydrocarbon (PAH) [3–7]. As a result of high airborne pollutant emissions from charcoal combustion, the Canadian government listed charcoal as a hazardous material [8]. According to the Canadian regulation

guidelines, the toxic fumes of charcoal may occur and cause death if the charcoal is burnt without adequate ventilation systems.

In this study, the emission characteristics of odorant pollutants were investigated from gases released from the combustion of charcoal products. To this end, the emission concentrations of key offensive odorants were measured from a total of 15 different barbecue charcoal products commercially available in the Korean market. In a number of previous studies, we investigated the emission characteristics of several pollutant groups including aromatic volatile organic compounds and carbonyls [4] and trace metals including mercury [6,7]. The metallic content in raw (unburnt) charcoal products was also investigated [5]. In an effort to extend the database of pollutant emissions from charcoal use, odorant emissions due to charcoal combustion were analyzed to unveil the basic features of this odorant nuisance in our everyday life activities.

2. Materials and methods

2.1. Sample collection

In order to assess emission concentration levels of odorant gases (Table 1) in charcoal fumes, a series of experiments were carried out by combusting 15 different charcoal products from 5 different countries (6 from Korea, 4 from China, 3 from Indonesia, 1 from Malaysia, and 1 from the US), all of which are commonly available in the S. Korean market. The collection of sample gases was made by following the procedures described in previous studies [4]. For the collection of gaseous samples, charcoal

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Table 1
List of odorous pollutants and reference components emitted from charcoal samples in this study.^a

Group	Order ^b	Full name	Short name	CAS Number	Chemical formula	Molecular weight (g mol ⁻¹)	Odor threshold (ppb)	Permissible concentration (ppb) ^c
VOC	1	Toluene	T	108-88-3	C ₇ H ₈	92.1	330	10,000
	2	p-Xylene	p-X	106-41-3	C ₈ H ₁₀	106.0	58	1000
	3	Styrene	S	100-42-5	C ₈ H ₈	104.0	35	400
	4	Methyl ethyl ketone	MEK	78-93-3	C ₆ H ₈ O	72.1	440	13,000
	5	Isobutyl alcohol	i-BuAl	78-83-1	C ₄ H ₁₀ O	74.1	11.00	900
	6	Methyl isobutyl ketone	MIBK	108-10-1	C ₆ H ₁₂ O	100.0	170	1000
	7	Butyl acetate	BuAc	123-86-4	C ₆ H ₁₂ O ₂	116.00	16	1000
RSC	8	Hydrogen sulfide	H ₂ S	7783-06-4	H ₂ S	34.1	0.41	20
	9	Methyl mercaptan	CH ₃ SH	74-93-1	CH ₃ SH	48.1	0.07	2
	10	Dimethyl sulfide	DMS	75-18-3	(CH ₃) ₂ S	62.1	3	10
	11	Dimethyl disulfide	DMDS	624-92-0	(CH ₃) ₂ S ₂	94.2	2.2	9
Aldehyde	12	Acetaldehyde	AA	75-07-0	C ₂ H ₄ O	44	1.5	50
	13	Propionaldehyde	PA	123-38-6	C ₃ H ₆ O	58.1	1	50
	14	Butyraldehyde	BA	123-72-8	C ₄ H ₈ O	72.1	0.67	29
	15	Isovaleraldehyde	IA	590-86-3	C ₅ H ₁₀ O	86.1	0.1	3
	16	Valeraldehyde	VA	110-62-3	C ₅ H ₁₀ O	86.1	0.41	9
N	17	Ammonia	NH ₃	7664-41-7	NH ₃	17	1500	1000
Relevant parameters	18	Formaldehyde	FA	50-00-0	CH ₂ O	30	500	-
	19	Benzene	B	71-43-2	C ₆ H ₆	78.1	2700	-
	20	Carbon disulfide	CS ₂	75-15-0	CS ₂	76.1	210	-
	21	Sulfur dioxide	SO ₂	7446-09-5	SO ₂	64.1	870	-
	22	Carbon monoxide	CO	630-08-0	CO	28	-	-
	23	Total hydrocarbon	THC	-	-	-	-	-
	24	Particulate matter (10 μg)	PM ₁₀	-	-	-	-	-

^a A total of five (trimethylamine and 4 volatile fatty acids (propionic acid, butyric acid, valeric acid, and isovaleric acid)) out of 22 offensive odorants were not considered; their concentrations were below detection limits from all samples.

^b Compounds of order 1–17 are offensive odorants designated by the malodor prevention law in Korea (KMOE, 2008) [13].

^c Permissible emission concentration of offensive odorants in non-industrial sectors according to the malodor prevention law in Korea (KMOE, 2008) [13].

products were combusted in an old-style Korean combustor (sample weight: ~550 g). Gases released from charcoal combustion were collected in 10 L Tedlar bags using a lung vacuum sampler (ACEN Co. Ltd., Korea). After releasing the first 5 min of gas emissions, gas samples were collected for 30 min into the Teflon bag placed in the lung sampler through a sampling line (made of Teflon) connected to the chimney of the combustor. These samples collected in the Tedlar bag were then used for the analysis of VOC and RSC.

For the collection of carbonyls, charcoal fumes were drawn into Lp-DNPH cartridges (Supelco Inc., USA) at a flow rate of 1 L min⁻¹ for 5 min via a Sep-Pak ozone scrubber (Waters, USA). In the case of ammonia (NH₃), gaseous samples were absorbed directly into a boric acid solution made by 2.5 g boric acid mixed into 500 mL. To induce absorption of NH₃, three impingers were connected to each other; two of them were filled with boric acid (20 mL each), while the other was filled with resin (for dehydration of outgoing air). Next, a vacuum pump (MP-Σ300, SIBATA, Japan) was used to draw gas samples through impinger at a flow rate of 2.5 L min⁻¹ to make a total of 30 L.

2.2. Analysis of odorant components

All collected samples were analyzed to determine concentration of up to 22 major offensive odorants designated by the malodor prevention law in Korea (KMOE, 2008) along with other reference pollutants (benzene, CS₂, SO₂, CO, THC, and PM₁₀) (Table 1). The concentration data of 5 of the 22 offensive odorants (trimethylamine, propionic acid, butyric acid, valeric acid, and isovaleric acid) were not evaluated due to their limited detectability. Instead, a list of reference compounds was selected in this study and analyzed due to their relative abundance, irrespective of their odorant intensities. The basic analytical settings and the basic experimental conditions for their analysis are described in Table 2. The analysis of these odorants was made by following the procedures introduced in a

number of previous investigations [5,9,10]. The basic information for these procedures is described briefly below.

For VOC analysis, the combination of gas chromatography (GC) with mass spectrometry (MS) coupled with a multifunction thermal desorber (TD) was used. The samples in the Tedlar bag were transported to the TD system for analysis based on the thermal desorption. Chromatographic separation was achieved by the Vocol column (60 m × 0.32 mm i.d. and 1.8-μm film thickness; Supelco) at a column flow rate 1.2 mL min⁻¹ (99.9% pure He as carrier gas). Detailed analytical conditions of this system are listed in Table 2. The detection limit (DL) values fell in the range of 1.27 (0.31 ppb (MIBK)) to 1.81 ng (0.38 ppb (BuAc)). If the precision of this method is evaluated in terms of relative standard error (RSE), it generally varied from 2.96% (MEK) to 4.96% (BuAc).

The analysis of RSC was made by GC with pulsed flame photometric detector (PFPD), which is interfaced with a multi-function TD and an air server (AS) unit. Detailed operating conditions of this system are described in Table 2. The analytical procedures of RSC can also be referred to in some of previous publications [11,12]. The DL values of RSC ranged from 0.5 pg (or 0.12 ppb (DMDS)) to 0.7 pg (or 0.52 ppb (H₂S)) in a sampling volume of 120 mL. Their RSC values ranged from 1.35 (H₂S) to 4.25% (DMDS).

For carbonyl analysis, high performance liquid chromatography (HPLC) equipped with a UV detector and dsCHROM software (for peak integration) was used. The basic analytical conditions of the HPLC system are provided in Table 2. The RSE values were in the range of 0.51% (AA) to 2.16% (IA). The DL of the carbonyls ranged from 0.04 (VA) to 0.52 ppb (FA). Furthermore, NH₃-absorbed boric acid was analyzed by the UV/VIS detector system based on the colorimetric method (Table 2). Quantification of NH₃ was carried out with a DL value of 133 ppb and RSE of 1.95%. Due to problems with sample handling, quantification of some VOCs (e.g., MEK, i-BuAl, MIBK, and BuAc) was confined to three Korean samples and one US sample (K1–K3 and U1) (Table 3). Apart from the target odorants, some reference components (PM₁₀, CO, and THC) were also analyzed in light of their abundance in fume samples.

Table 2
Experimental conditions of the instrumental systems used for odorant detection in this study.

[1] GC/MS system for VOC analysis				
1. GC/MS (SHIMADZU GCMS-QP2010, Japan)				
(a) Oven condition				
Initial temp.:	35	°C	(b) Detector (MS)	
Hold time:	4	min	Ionization mode:	EI (70 eV)
Ramping rate:	4	°C min ⁻¹	Ion source temp.:	200 °C
Final temp.:	200	°C	TIC scan range:	35–250 m/z
Hold time:	10	min	Threshold:	100
Carrier gas:	He	99.90%		
(c) Column (Vocol, PA, USA)				
			Column (Vocol, PA, USA)	0.32 mm
			Length:	60 m
			Film thickness:	1.8 μm
2. Thermal desorber (UNITY, Markers International Ltd., UK)				
Cold trap:	Carbopack B+ Tenax		Trap low:	5 °C
Split ratio:	20		Trap high:	300 °C
Split flow:	5.0	mL min ⁻¹	Flow path temperature:	120 °C
Hold time:	5.0	min		
[2] GC/TD system for RSC analysis				
1. GC/PFPD (DS 6200, Donam Instrument, Korea) system				
(a) Oven condition				
Initial temp.:	80	°C	(b) Detector (PFPD: Model 5380, O.I. Analytical, USA)	
Ramping rate:	20	°C min ⁻¹	Detector temp.:	250 °C
Final temp.:	200	°C	Air(1)/air(2): flow:	10 mL min ⁻¹
Initial hold:	4.5	min	H ₂ flow:	11.5 mL min ⁻¹
Final hold:	9.5	min		
Total time:	20	min	(c) Column (BP-1, SGE, Australia)	
2. Thermal desorber (UNITY, Markers International Ltd., UK)				
Cold trap:	Carbopack B+ Silica gel = 1.5:2.5		Film thickness:	5 μm
Split ratio:	10:01		Length:	60 m
Split flow:	15	mL min ⁻¹	Diameter:	0.32 mm
Hold time:	5	min	Trap low:	-15 °C
			Trap high:	250 °C
			Flow path temperature:	80 °C
[3] HPLC (Series 1500, Lab Alliance, USA)/UV system for carbonyl compounds analysis				
Injector				
Volume:	20	μL	Column (C18, Hichrom, UK)	
Pump				
Flow rate:	1.5	mL min ⁻¹	Column dimensions:	250 × 46 mm
Mobile phase:	Acetonitrile:water	70:30	Particle size:	5 μm
Analysis time:	15	min	Pore size:	300 Å
UV detector (Model 500, Lab Alliance, USA)				
Wavelength:	360	nm	Temp.:	20 °C
			Temp.:	Monomeric
[4] UV/VIS Spectrometer (Genesys™ 10 series, Thermo Electron Corp., USA) system for ammonia analysis				
Impringer system				
Pump flow rate:	2.5	mL min ⁻¹	Detector:	UV/VIS
Volume absorbed:	30	L	Wavelength:	635 nm
Absorption time:	12	min		
Temperature:	22	°C		
Boric acid volume:	50	mL		

3. Results and discussion

3.1. General features of odorant pollutant emission during combustion

The emission concentrations of odorant pollutants with diverse chemical properties were measured from 15 different charcoal products made in five different countries (Table 3). A brief inspection of the data indicates that the magnitude of odorant emissions differs significantly between compounds and between individual charcoals. According to the statistical summary provided in Table 4, the mean values of AA, PA, IA, and ammonia exceeded the guideline of malodor prevention law in Korea [13]. Their mean concentrations were almost 20 (1009 ppb), 7 (354 ppb), 2 (6.35 ppb), and 7 times (7800 ppb) higher than their respective guideline values. In contrast, emissions of certain species (RSCs, VA, etc.) were commonly limited in their detectability in most samples (Table 3).

Although VOCs are the key components of charcoal combustion, their concentrations are greatly distinguished between samples.

Among VOCs, p-X was measured consistently above DL in all charcoal products except for the one made in the US. Although benzene is not an odorant, its concentration levels from glowing charcoals have been measured frequently [14]. Due to the carcinogenic effect and the susceptibility of inhalation [15], benzene is designated as one of the top-priority compounds for the development of indoor air quality guidelines [16]. In this study, the concentration (ppb) of benzene averaged at 843, which was far higher than toluene (4.07). This contrasts with a comparable trend between toluene (116) and benzene (98.7) in previous study [4]. In the case of RSCs, their emission concentrations were generally low compared to VOCs and aldehydes. Although odorant RSCs were seen abundantly from a Korean sample (K2), they were not measured from any other sample. In contrast, the emission concentrations of other sulfur species (CS₂ and SO₂) from our samples were relatively high with averages of 3.85 and 14.9 ppb, respectively. In this research, a total of five odorant carbonyl (AA, PA, BA, IA, and VA) were measured along with FA as a reference. Among aldehydes, the concentrations (ppb) of AA (1009) and FA (354) were generally the highest, while the

Table 3
Emission concentrations (ppb) of target odorants and reference compounds measured from charcoal combustion experiments in this study.

Group	Order	Short name ^a	Sample code ^b																	
			K1	K2	K3	K4	K5	K6	C1	C2	C3	C4	I1	I2	I3	M1	U1			
VOC	1	T	21.8	14.1	15.5	0.94	<u>0.0003</u> ^c	<u>0.0003</u>	<u>0.0003</u>	<u>0.0003</u>	<u>0.0003</u>	<u>0.0003</u>	<u>0.0003</u>	<u>0.0003</u>	<u>0.0003</u>	<u>0.0003</u>	<u>0.0003</u>	0.65	8.10	
	2	p-X	0.44	492	3.10	1.63	27.00	4.39	1.09	1.07	1.17	1.69	1.58	1.21	0.39	0.94	0.08	0.08	0.08	
	3	S	6.73	7629	57.7	<u>0.0005</u>	10.0	<u>0.0005</u>	<u>0.0005</u>	<u>0.0005</u>	<u>0.0005</u>	<u>0.0005</u>	<u>0.0005</u>	<u>0.0005</u>	<u>0.0005</u>	<u>0.0005</u>	<u>0.0005</u>	<u>0.0005</u>	<u>0.0005</u>	<u>0.0005</u>
	4	MEK	5.74	5915	108	–	–	–	–	–	–	–	–	–	–	–	–	–	–	0.17
	5	i-BuAl	<u>0.26</u>	2001	0.26	–	–	–	–	–	–	–	–	–	–	–	–	–	–	0.17
	6	MIBK	<u>0.17</u>	<u>0.17</u>	2.36	–	–	–	–	–	–	–	–	–	–	–	–	–	–	0.08
	7	BuAc	<u>0.29</u>	<u>0.29</u>	<u>0.29</u>	–	–	–	–	–	–	–	–	–	–	–	–	–	–	0.08
RSC	8	H ₂ S	<u>0.03</u>	3.38	<u>0.03</u>	<u>0.036</u>	<u>0.036</u>	<u>0.036</u>	<u>0.036</u>	<u>0.036</u>	<u>0.036</u>	<u>0.036</u>	<u>0.036</u>	<u>0.036</u>	<u>0.036</u>	<u>0.036</u>	<u>0.036</u>	<u>0.036</u>	<u>0.036</u>	<u>0.25</u>
	9	CH ₃ SH	<u>0.03</u>	14.4	<u>0.03</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.23</u>
	10	DMS	<u>0.02</u>	97.5	0.24	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.024</u>	<u>0.17</u>
Aldehyde	11	DMDS	0.09	42.4	0.14	<u>0.017</u>	<u>0.017</u>	<u>0.017</u>	<u>0.017</u>	<u>0.017</u>	<u>0.017</u>	<u>0.017</u>	<u>0.017</u>	<u>0.017</u>	<u>0.017</u>	<u>0.017</u>	<u>0.017</u>	<u>0.017</u>	<u>0.017</u>	<u>0.11</u>
	12	AA	83.0	5764	1415	213	5734	189	883	47.1	68.4	110	198	241	107	81.4	0.44	0.44	0.44	
	13	PA	5.07	347	80.91	<u>0.05</u>	388	<u>0.05</u>	<u>0.05</u>	<u>0.05</u>	<u>0.05</u>	<u>0.05</u>	<u>0.05</u>	<u>0.05</u>	<u>0.05</u>	14.1	<u>0.05</u>	<u>0.05</u>	<u>0.05</u>	<u>0.44</u>
	14	BA	<u>0.46</u>	89.1	71.6	<u>0.05</u>	132	12.3	<u>0.05</u>	<u>0.05</u>	<u>0.05</u>	<u>0.05</u>	<u>0.05</u>	<u>0.05</u>	<u>0.05</u>	<u>0.05</u>	<u>0.05</u>	<u>0.05</u>	<u>0.05</u>	<u>0.45</u>
	15	IA	<u>0.47</u>	47.6	21.5	<u>0.04</u>	24.7	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.56</u>
	16	VA	<u>0.48</u>	21.4	0.48	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.04</u>	<u>0.49</u>
	17	NH ₃	22,319	83,325	8928	<u>21</u>	<u>55</u>	<u>24</u>	<u>18</u>	<u>10</u>	<u>24</u>	<u>26</u>	<u>16</u>	<u>3</u>	<u>5</u>	<u>9</u>	<u>1614</u>	<u>1614</u>	<u>1614</u>	<u>1614</u>
Relevant parameters	18	FA	99.4	13.6	831	397	1137	436	138	174	198	299	194	902	153	340	0.52	0.52	0.52	
	19	B	<u>0.03</u>	6,535	221	132	2093	1081	314	98.8	325	254	100	1120	59.3	283	21.7	21.7	21.7	
	20	CS ₂	2.07	40.0	0.15	1.84	3.34	1.22	1.43	0.95	0.65	1.52	1.54	0.94	0.53	1.44	0.07	0.07	0.07	
	21	SO ₂	39.7	21.9	8.95	6.48	20.5	9.35	2.51	12.1	26.0	14.9	4.43	37.6	0.83	3.54	–	–	–	
	22	CO	1,020,000	1,050,000	537,000	399,375	3,612,038	1,316,450	1,662,743	999,473	2,621,175	1,838,183	901,390	1,727,745	1,309,458	1,525,533	750	750	750	
	23	THC ^d	7300	87,200	<u>500</u>	212,760	2,034,143	1,730,000	169,093	<u>500</u>	199,360	161,325	<u>500</u>	983,400	<u>500</u>	84,280	<u>500</u>	<u>500</u>	<u>500</u>	
	24	PM ₁₀ ^e	800	8889	24,889	3833	2833	3667	2000	2667	2667	2667	2000	2500	3000	500	717	717	717	

^a Refer to Table 1 for full name.

^b Sample code: K (Korea); C (China); I (Indonesia); M (Malaysia); and U (US).

^c Underlined numbers denote concentration values below detection limit. DL values of a given species are occasionally different due to measurement intervals or changes in sample loading condition or sample treatments. Superscript letters d and e denote concentration units of ppbC and $\mu\text{g m}^{-3}$, respectively.

Table 4
Comparison of mean concentration values of odorants and reference components measured from 15 BBQ charcoal products.

Group	Order	Short name ^b	All	Korea	China	Indonesia	Malaysia	US
A. Offensive odorants (unit: ppb) ^a								
VOC	1	T	4.07	8.71	0.0003 ^c	0.0003	0.65	8.10
	2	p-X	35.9	88.2	1.26	1.06	0.94	0.08
	3	S	514	1284	0.21	0.0005	0.0005	0.06
	4	MEK	1507	2009	–	–	–	0.17
	5	i-BuAl	500	667	–	–	–	0.17
	6	MIBK	0.64	0.84	–	–	–	0.08
	7	BuAc	0.12	0.29	–	–	–	0.08
RSC	8	H ₂ S	0.25	0.58	0.036	0.036	0.036	0.25
	9	CH ₃ SH	0.98	2.41	0.024	0.024	0.024	0.23
	10	DMS	6.53	16.3	0.024	0.024	0.024	0.17
	11	DMDS	2.85	7.11	0.017	0.017	0.017	0.11
Aldehyde	12	AA	1009	2233	277	182	81.40	0.44
	13	PA	55.7	137	0.05	4.72	0.05	0.44
	14	BA	20.4	50.9	0.05	0.05	0.05	0.45
	15	IA	6.30	15.7	0.04	0.04	0.04	0.56
	16	VA	1.49	3.65	0.04	0.04	0.04	0.49
N	17	NH ₃	7753	19,104	19.7	8.1	9.0	1614
B. Relevant parameters (unit: ppb)								
	18	FA	354	486	202	416	340	0.26
	19	B	843	1677	248	426	283	21.7
	20	CS ₂	3.85	8.11	1.14	1.0	1.44	0.03
	21	SO ₂	14.9	17.8	13.87	14	3.54	–
	22	CO	1,368,062	1,322,477	1,780,393	1,312,864	1,525,533	375
	23	THC ^d	378,007	678,609	132,507	327,967	84,280	250
	24	PM ₁₀ ^e	4242	7485	2500	2500	500	717

^a In case all data for a given country are below detection limit (BDL), it was used directly for the computation of mean value; however, if the BDL data are mixed with the normal data, then one half of BDL was used to derive the mean value for each country.

^{b,c} Refer to Tables 1 and 3, respectively.

^d Concentration unit = ppbC.

^e Concentration unit = $\mu\text{g m}^{-3}$.

detection of other carbonyls (PA, BA, IA, and VA) were generally confined in most samples. Moreover, fairly high concentrations of ammonia were recorded from many samples (such as K-2, -1, -3, and U1), which were around 83, 22, 10 and 1.5 times higher than the guidelines [13]. As ammonia undergoes gas-to-particle conversion with atmospheric acids, it may be present in salt form in the atmosphere [17,18].

The concentrations of the major reference components (CO, THC, and PM₁₀) were measured consistently in most samples with the mean of 1368 ppm, 378 ppmC, and 4242 $\mu\text{g m}^{-3}$, respectively (Table 4). As the occupational exposure of CO is generally regulated in the range of 20 to 50 ppm [19,20], its value measured in this study was about 27–68 times higher than those guideline values. Hence, if ventilation is not maintained properly, charcoal combustion may cause severe health effects. As such, the prevalence of high PM₁₀ levels from charcoal combustion is also a worrying component [21].

3.2. Comparison of odor strengths between different charcoal products in terms of odor intensity

As odors released either directly or indirectly from human activities cause adverse health effects, they are often classified as the contaminants requiring regulation [22]. In North America and Europe, odors have been ranked as the major targets of public complaints to regulatory agencies [23]. Due to the complexity of odorant mixing and/or the delicacy of its detection by the human nose, a quantitative description of odor exposure is limited [24]. For this reason, the use of the odor intensity (OI) concept is an efficient approach as it provides a parallel means to evaluate concentration data of the perceived odorants through numerical conversions [9].

For the conversion of odorant concentrations into the OI values, empirical equations introduced by Nagata [25] were used in our study. The OI scaling of 0–6 can be distinguished as follows: 0 (no odor), 1 (very weak), 2 (weak), 3 (distinct), 4 (strong), 5 (very strong), and 6 (intolerable) [26]. However, as the OI values

were converted to negative values in some cases, such values were excluded from comparative analysis for the sake of simplicity. The OI values of offensive odorants derived via such conversions are compiled in Table 5.

In summary, our data suggests that the magnitude of OI values differs greatly among different charcoal products. If the OI values are compared between different charcoal products, aldehydes were found to be the most dominant odorant followed by VOC. Although FA and AA were detected abundantly from most charcoal samples, other carbonyls (PA, BA, and IA) were only found abundantly from 3 Korean samples e.g., (K-2, 3, and 5). It should be noted that MIBK and BuAc were not significant as an odorant, with OI values below zero. In contrast, NH₃ exhibited the highest OI value (5.58) in K-2 charcoal, while its values for K-1, -3, and U1 were 4.63, 3.97, and 2.72, respectively.

As a simple means to assess the overall contribution to odor formation by all target compounds, the OI values of each individual compound were bound together to derive the total odor strength in terms of the “sum of odor intensity (SOI)”. For the derivation of the SOI term, the following equations were employed in this study [27]:

$$\text{SOI} = \log(\sum 10^{\text{OI}(\text{ith})}) = \log(10^{\text{OI}(\text{ith})1} + 10^{\text{OI}(\text{ith})2} + 10^{\text{OI}(\text{ith})3} + \dots + 10^{\text{OI}(\text{ith})n})$$

where $\text{OI}(\text{ith}) = \log 10^{\text{OI}(\text{ith})}$.

As the mixing of odorants is a complicated process, one should bear in mind that the derivation of SOI should be considered an arbitrary approach to assess one of the various mixing effects. In terms of SOI, the strength of the odor emission peaked in K-2 (5.66) followed by K-5 (4.73), K-1 (4.61), K-3 (4.43), C-1 (3.80). In the rest of the charcoal products, SOI values ranged from 3.14 to 2.51 (Table 5). In terms of odor strength classification into a 0–6 numerical scale by the American Society for Testing and Materials [26], the result of different charcoal products was arbitrarily

Table 5
Comparison of odor intensity (OI) for each individual charcoal product investigated in this work.

Order	Compound	Function ^a	Sample code ^b																
			K1	K2	K3	K4	K5	K6	C1	C2	C3	C4	I1	I2	I3	M1	U1		
1	T	$Y = 1.380 \log X + 4.60$	2.31	2.05	2.10	0.42	-	-	-	-	-	-	-	-	-	-	-	1.71	
2	p-X	$Y = 1.570 \log X + 2.44$	- ^c	1.96	-	-	-	-	-	-	-	-	-	-	-	-	-	-	
3	S	$Y = 0.790 \log X + 2.53$	0.81	3.23	1.55	-	-	-	-	-	0.09	-	-	-	-	-	-	-	
4	MEK	$Y = 1.850 \log X + 0.15$	-	1.58	-	-	-	-	-	-	-	-	-	-	-	-	-	-	
5	i-BuAl	$Y = 0.790 \log X + 2.53$	-	2.77	-	-	-	-	-	-	-	-	-	-	-	-	-	-	
6	MIBK	$Y = 1.650 \log X + 2.27$	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	
7	BuAc	$Y = 1.140 \log X + 2.34$	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	
8	H ₂ S	$Y = 0.950 \log X + 4.14$	-	1.79	-	-	-	-	-	-	-	-	-	-	-	-	-	-	
9	CH ₃ SH	$Y = 1.250 \log X + 5.99$	-	3.69	-	-	-	-	-	-	-	-	-	-	-	-	-	-	
10	DMS	$Y = 0.784 \log X + 4.06$	-	3.27	1.22	0.44	-	-	-	-	-	-	-	0.44	-	-	-	-	
11	DMDS	$Y = 0.985 \log X + 4.51$	-	3.16	0.72	-	-	-	-	-	-	-	-	-	-	-	-	-	
12	AA	$Y = 1.010 \log X + 3.85$	2.76	4.62	4.00	3.17	3.12	3.80	2.51	2.67	2.88	3.14	3.23	2.87	2.75	-	-	-	
13	PA	$Y = 1.010 \log X + 3.86$	1.53	3.40	2.76	-	-	-	-	-	-	-	-	-	-	-	-	-	
14	BA	$Y = 1.060 \log X + 4.23$	-	3.12	3.02	-	-	-	-	-	-	-	-	-	-	-	-	-	
15	IA	$Y = 1.350 \log X + 6.01$	-	4.22	3.76	-	-	-	-	-	0.07	0.07	-	-	-	-	-	-	
16	NH ₃	$Y = 1.670 \log X + 2.38$	4.63	5.58	3.97	-	-	-	-	-	-	-	-	-	-	-	-	2.72	
17	FA	$Y = 1.530 \log X + 1.59$	0.06	-	1.47	0.98	1.04	0.27	0.43	0.51	0.79	0.50	1.52	0.34	0.87	-	-	-	
	SOI		4.61	5.66	4.43	3.17	3.17	3.80	2.51	2.67	2.88	3.14	3.24	2.92	2.76	-	-	2.76	

^a Nagata [25], odor intensity (Y) and odorant concentration (ppm) (X).

^b Sample code: K (Korea); C (China); I (Indonesia); M (Malaysia); and U (US).

^c Negative OI values are not considered.

divided into 4 groups by their magnitude (1) Very strong (K-2), (2) Strong (K-1, K-3, and K-5), (3) Distinct (K-4, K-6, C-1, I-1, and I-2), and (4) Weak for all the remaining charcoal. According to a compilation of the SOI values between different countries, the odor intensity of the Korean samples tends to be significantly high compared to others (e.g., Malaysia and US).

3.3. Comparison of odorants between charcoal products of different countries

As our measurements were made to explore odorant emission characteristics covering 15 products from five different countries, the data can be evaluated to assess the possible role of such criteria as the origin of the charcoal production. As seen by the plot of the averaged concentrations from all the experimental results, charcoal consumption appears to be a significant odorant emission process (Fig. 1). If the mean values of the major odorants are compared between countries, the values of the Korean charcoal products record the highest of all countries (Fig. 2). In many Korean charcoals, the concentration values of all odorants (except T, p-X, MEK, and DMDS, Table 5) were high enough to exceed their respective guideline emission values [e.g., 13]. If we look into the common trend of pollutant emissions between different countries (such as Korea, Chinese, Indonesia, and Malaysia samples), the acetaldehyde (AA) value was about 45, 6, 4, and 2 times higher than the Korean guideline value. In the case of ammonia, it was found in a very high concentration range in Korean and US charcoal; its values were about 20 and 2 times higher than the guideline value (Table 4). It is worth noting that ammonia was the only prime odorant released from the US charcoal product tested in this study, while it was not for the products of the other countries.

If we compare the odor strengths of charcoal products between different countries in terms of SOI values, Korean products recorded the highest SOI value of 4.30 followed by Indonesian (3.10), Chinese (2.97), Malaysian (2.76), and the US product (2.76) (Fig. 2). The SOI values of the Korean charcoal products showed a little complicated pattern between different odorants, while those of other countries (Chinese, Indonesian, and Malaysian charcoal products) appear to be dominated by the AA compound. However, the pattern of US products is quite exceptional, as ammonia was the single dominant odorant contributing to the SOI level. This observation thus suggests the possibility that such product underwent a sufficient cleaning process in the handling of the raw materials compared to other countries.

3.4. Factors affecting odorant emissions from charcoal

In order to learn more about the factors affecting the odorant emissions from various charcoal products, Pearson's correlation analysis was done using the concentration data of all odorants (Table 6). Concentrations of some odorants (MEK, i-BuAl, MIBK, H₂S, CH₃SH, DMS, IA, and VA) were excluded in the correlation analysis due to the limited detectability. If the results are compared by the level of statistical significance (*p* value) of 0.05 and 0.01, a total of 6 and 28, respectively, out of 94 matching pairs were found to be statistically significant. In our study, benzene displayed a good correlation with most of the compounds, while toluene and p-X were only correlated significantly with ammonia. Among the aldehydes, FA, AA, PA, and BA showed good correlations with each other, suggesting the similar nature of their source processes among different carbonyls. In most cases, odorants of VOC (P, p-X, and S) were highly correlated with aldehydes (AA, PA, and BA).

In order to assess the source of different odorants (and pollutants) in charcoal fume, both the manufacturing process and raw materials used for its production need to be investigated. The natural raw materials for charcoal production are based on wood, both

Table 6

Results of correlation analysis between target odorants investigated and reference components in this study.^a

	T	PX	S	DMDS	AA	PA	BA	NH ₃	FA	B	CS ₂	SO ₂	CO	THC	PM ₁₀
(a) Results															
T	<i>r</i> ^b	1													
	<i>p</i> ^c	-													
	<i>N</i> ^d	15													
PX	<i>r</i>	0.376													
	<i>p</i>	0.168	1												
	<i>N</i>	15	15												
S	<i>r</i>	0.387	0.999**	1											
	<i>p</i>	0.154	0.000	-											
	<i>N</i>	15	15	15											
DMDS	<i>r</i>	0.386	0.999**	0.999**	1										
	<i>p</i>	0.155	0.000	0.000	.										
	<i>N</i>	15	15	15	15										
AA	<i>r</i>	0.220	0.708**	0.672**	0.671**	1									
	<i>p</i>	0.431	0.003	0.006	0.006	-									
	<i>N</i>	15	15	15	15	15									
PA	<i>r</i>	0.220	0.666**	0.629*	0.627**	0.992**	1								
	<i>p</i>	0.432	0.007	0.012	0.012	0.000	.								
	<i>N</i>	15	15	15	15	15	15								
BA	<i>r</i>	0.268	0.497	0.459	0.456	0.930**	0.944**	1							
	<i>p</i>	0.334	0.059	0.085	0.088	0.000	0.000	.							
	<i>N</i>	15	15	15	15	15	15	15							
NH ₃	<i>r</i>	0.608 [†]	0.958**	0.962**	0.962**	0.633 [†]	0.595 [†]	0.449	1						
	<i>p</i>	0.016	0.000	0.000	0.000	0.011	0.019	0.093	.						
	<i>N</i>	15	15	15	15	15	15	15	15						
FA	<i>r</i>	-	-	-	-	0.327	0.358	0.541*	-	1					
	<i>p</i>	0.550	0.388	0.325	0.319	0.234	0.190	0.037	0.290	.					
	<i>N</i>	15	15	15	15	15	15	15	15	15					
B	<i>r</i>	0.263	0.954**	0.940**	0.940**	0.829**	0.794**	0.643**	0.880**	0.004	1				
	<i>p</i>	0.344	0.000	0.000	0.000	0.000	0.000	0.010	0.000	0.987	.				
	<i>N</i>	15	15	15	15	15	15	15	15	15	15				
CS ₂	<i>r</i>	0.374	0.998**	0.997**	0.997**	0.707**	0.664**	0.487	0.960**	-	0.952**	1			
	<i>p</i>	0.170	0.000	0.000	0.000	0.003	0.007	0.066	0.000	0.375	0.000	.			
	<i>N</i>	15	15	15	15	15	15	15	15	15	15	15			
SO ₂	<i>r</i>	0.437	0.165	0.159	0.160	0.163	0.186	0.124	0.293	0.190	0.243	0.173	1		
	<i>p</i>	0.119	0.573	0.587	0.585	0.579	0.525	0.672	0.309	0.515	0.403	0.554	.		
	<i>N</i>	14	14	14	14	14	14	14	14	14	14	14	14	14	
CO	<i>r</i>	-	-	-	-	0.408	0.430	0.397	-	0.483	0.153	-	0.280	1	
	<i>p</i>	0.131	0.825	0.724	0.722	0.131	0.110	0.143	0.566	0.068	0.586	0.852	0.333	.	
	<i>N</i>	15	15	15	15	15	15	15	15	15	15	15	15	15	
THC	<i>r</i>	-	-	-	-	0.399	0.424	0.471	-	0.689	0.209	-	0.200	0.603 [†]	1
	<i>p</i>	0.244	0.772	0.664	0.662	0.140	0.115	0.076	0.519	0.005	0.456	0.779	0.493	0.017	.
	<i>N</i>	15	15	15	15	15	15	15	15	15	15	15	15	15	15
PM ₁₀	<i>r</i>	0.456	0.214	0.220	0.215	0.264	0.248	0.480	0.266	0.348	0.185	0.184	-	-	1
	<i>p</i>	0.087	0.443	0.431	0.441	0.341	0.372	0.070	0.338	0.204	0.510	0.511	0.691	0.385	0.678
	<i>N</i>	15	15	15	15	15	15	15	15	15	15	15	14	15	15

(b) Summary of correlation analysis

Sample source	Frequency of matching pairs at 2 significance levels		Total number of possible matching pairs
	0.01	0.05	
	28	6	

^a A total of 9 odorants (MEK, i-BuAl, MIBK, BuAc, H₂S, CH₃SH, DMS, IA, and VA) 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.

* Correlation is significant at the 0.05 level (2-tailed).

** Correlation is significant at the 0.01 level (2-tailed).

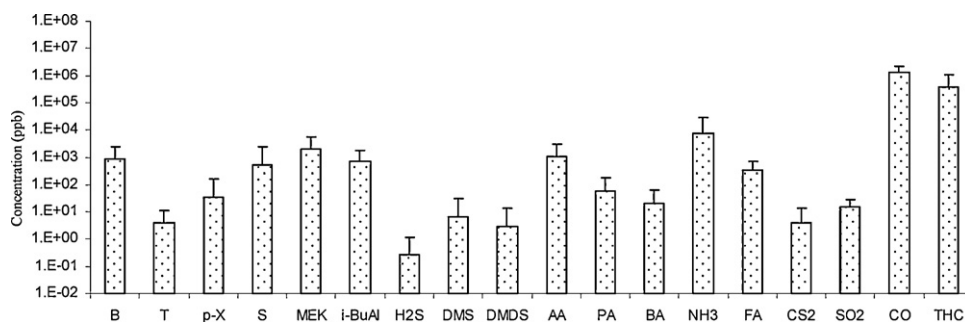


Fig. 1. Comparison of the mean concentration of major odorants and reference components measured from 15 different charcoal products (unit of THC = ppbC).

hardwood and softwood. In addition, sawdust, wood shavings, fruit stones, nuts, nutshells, corn cobs, bark, cotton seeds, and similar products can also be employed. Supply of wood came from two sources, directly from the forest or from industrial waste [28]. In light of this fact, it is logical to infer that recycled industrial wood, if used for charcoal, may play a certain role in the emission of hazardous odorants from charcoal fumes. Moreover, in our investigation, few charcoal products produced significant amounts of tarry residue during combustion. Note that liquid and tarry residues were not fully removed by the process of carbonization during production [29]. In addition, according to an investigation by the US EPA, when pyrolysis is used to manufacture charcoal from wood, a variety of different organic compounds are released into the air, depending on the specific pyrolysis or burn conditions [30]. The content of volatile components is almost zero, if carbonization is achieved at high temperatures (around 1000 °C). However, the negative effect of such processes is that the production can fall about 25% compared to low temperature carbonization [31]. In light of evidence collected from several previous researches, it is evident that the notable differences in pollutant emission levels were affected by the combined effects of their manufacturing ingredients and processes [4,5].

3.5. Assessment of potential health risks of odorants from charcoal combustion

The smoke from biomass fuels (wood, agricultural waste, and dung) is an important source of indoor air pollution because large amounts of smoke inhalation may induce a health risks of a similar order of magnitude as the risk from tobacco smoke [32]. In

this study, major hazardous pollutants from charcoal combustion (odorants) were identified to include toluene, xylene, styrene, formaldehyde, acetaldehyde, and ammonia. According to [33], such compounds have the potential to pose cancer and non-cancer risks when present in dwellings and public buildings.

Most of the aromatics seen abundantly in this study (toluene, xylene, and styrene) are known to cause adverse health effects such as fatigue, sleepiness, headache, throat irritation, and short term memory loss [34]. During recent research on styrene, it was also found that its inhalation at both high and low concentration levels can damage DNA repair genes and pose long term cancer risks [35]. Although the level of odorant RSCs was insignificant, considerable amounts of other sulfur species like CS₂ and SO₂ were found in our study. In many previous studies, the inhalation of CS₂ is reported to destabilize the mechanism of carbohydrate metabolism in the human body [36–38]. In addition, SO₂ is known to exert certain effects on pulmonary function even at low concentrations [39].

A number of aldehydes, especially FA, AA, and IA, were frequently found to extend the guidelines of KMOE [13]. According to the toxic release inventory (TRI) data of the US EPA, acetaldehyde is also classified as a known or suspected carcinogen [40,41]. The reference compound, formaldehyde (>100 ppb) is also well known to cause irritation of eyes, nose, and throat, while being capable of accelerating asthma symptoms and other respiratory illnesses [42–44].

In addition, the primary acute effect of inhalation exposure to acetaldehyde includes irritation of the eyes, skin, and respiratory tract in humans, while at higher exposure levels, erythema, coughing, pulmonary edema, and necrosis may also occur [45]. In this

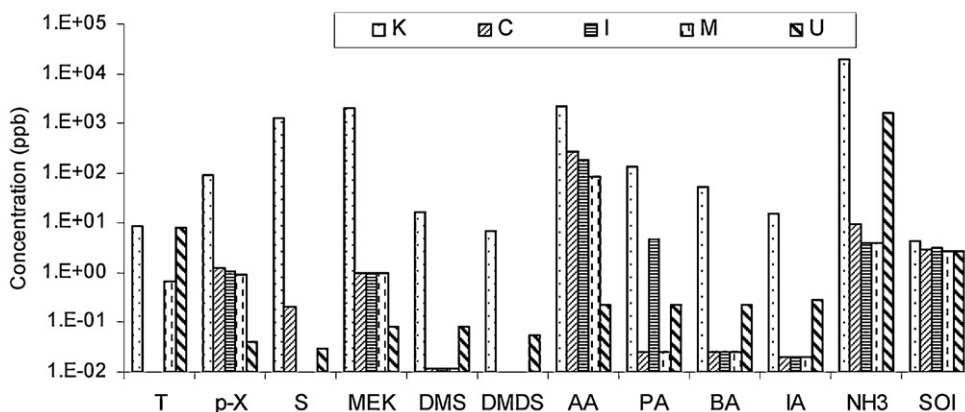


Fig. 2. Comparison of odorant concentrations between different countries of charcoal production (single letter symbol for country code).

investigation, we found several charcoals yielded a high concentration of ammonia which exceeded the guideline levels [13]. A recent study revealed that ammonia gas can cause damage, when hydrous ammonia reacts with tissue water to form strongly alkaline solution, ammonium hydroxide. It is a product from an exothermic reaction and can cause significant thermal injury to the skin, eyes, and especially to the respiratory system [46].

4. Conclusion

According to this study, several odorants (toluene, p-X, S, MEK, AA, PA, IA, and NH₃) are often significantly emitted during charcoal combustion and thereby play an important role in odor pollution under certain circumstances. Although emissions of many odorants from different origins were below harmful levels, a few odorants (AA, PA, IA, and NH₃) were found to exceed certain guidelines set for odorant emission control. If the strength of odor emission is compared in terms of sum of odor intensity (SOI), the values varied in the range of 2.51 to 5.66. As such, the emission levels of odorants from some charcoal products were significant enough to cause strong odor pollution. An evaluation of odorant levels released from charcoal combustion suggests the possibility that the materials used for charcoal production may play a vital role in terms of odor emission. Apart from odorants, our analysis confirmed that emission of other health hazardous reference components (benzene, CS₂, SO₂, CO, THC, and PM₁₀) can occur considerably from charcoal combustion. Given this situation, efforts should be made to properly establish regulation guidelines for charcoal production with respect to odor as well as other airborne pollutant emission. The results of a certain charcoal product like the one product made in the US suggest the possibility that odorant emission levels can be significantly reduced with proper treatment (during production). While seeking safer products for use, due to hazardous health effects especially in indoor environments, proper ventilation should also be implemented to reduce adverse health impacts from charcoal use.

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