



# Barbecue charcoal combustion as a potential source of aromatic volatile organic compounds and carbonyls

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## ABSTRACT

The emission concentrations of a number of aromatic volatile organic compounds (VOCs) and carbonyl compounds were quantified during the combustion of commonly used barbecue charcoal. The concentrations of VOC and carbonyls were determined by gas chromatography coupled with thermal desorption and HPLC method, respectively. The analysis of VOC emission concentrations showed that toluene ( $116 \pm 444$  ppb) was the most abundant. On the other hand, the carbonyls were dominated by formaldehyde ( $275 \pm 477$  ppb) and acetaldehyde ( $126 \pm 229$  ppb). A line of evidence indicates that the emission patterns of these pollutants are associated with the diverse nature of raw materials and the processes involved in their production. Although emission concentrations of target compounds were in most cases below the permissible exposure limits (PEL), a proper regulation against the use of BBQ charcoal is needed to reduce potential health risks associated with its use.

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## 1. Introduction

Charcoal is widely used for barbecuing, as it is a more manageable means for cooking than wood fire. It is also advantageous in generating more heat with less smoke than wood in the same quantity [1]. Although barbecues have usually been performed outdoors, they have also been applied indoors. As charcoal is burnt at high temperatures during the barbecuing process, the pollutants emitted from charcoal combustion can be absorbed in food and degrade air quality in the surrounding environment. The people nearby are thus exposed to pollutants with potential health risks and are apt to inhale emission gases [2].

In some countries such as Korea, barbecued meats are very popular. Hence, there are many barbecue-style restaurants in Korea which use charcoal burners. Degradation of indoor air by BBQ activities can be reflected in increases in respirable suspended particulates (RSP), carbon monoxide (CO), and other airborne pollutants at trace quantities (such as volatile organic compounds (VOCs) and carbonyl compounds) [3]. Both VOCs and carbonyls can be classified as common airborne contaminants emitted from fried food on a hot steel pan or broiling food on steel bars above the charcoal burner [4].

VOCs like benzene cause irritation to the eyes, nose, and throat, headaches, loss of coordination, nausea, damage to liver, kidney and central nervous system, etc. [5]. Common symptoms from acute exposure to carbonyl compounds are eye and skin burns, irritation of the respiratory system, mood swings, and nausea [6]. Because of health hazards, some carbonyl compounds like acetaldehyde are considered carcinogenic [7]. As such, numerous VOCs and carbonyls simultaneously constitute an important component in the assessment of health hazards and the impact of air quality degradation [8,9].

In barbecue restaurants, the release of indoor pollutants may be greatly stimulated, as customers cook their food over heated charcoal placed on customers' tables [10–12]. Nonetheless, there have been few studies intended to characterize the airborne pollution associated with the combustion of barbecue charcoal. The objective of this study is to determine and compare the carbonyl and VOC emissions from charcoal combustion. Our initial study on this subject focused on the emission of volatile mercury, and the results were reported elsewhere [2].

## 2. Materials and methods

### 2.1. Sample collection

In this study, target analytes were selected to cover common airborne organic pollutants grouped as aromatic VOC and carbonyls (Table 1). To assess the basic features of VOC and carbonyl emissions

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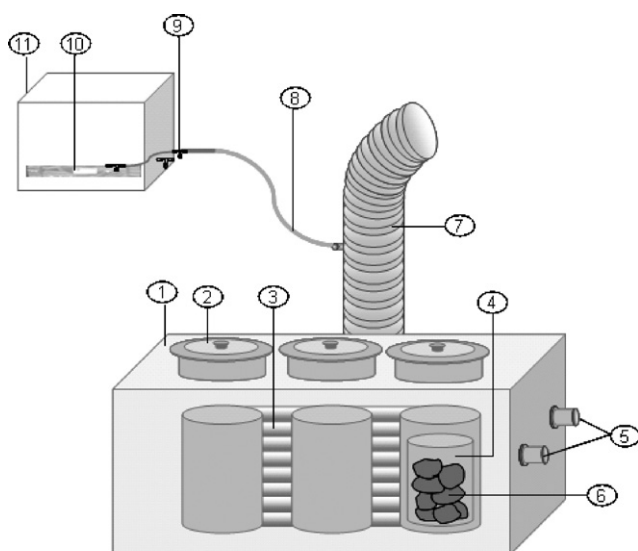
E-mail addresses: [khkim@sejong.ac.kr](mailto:khkim@sejong.ac.kr), [kkim61@nate.com](mailto:kkim61@nate.com) (K.-H. Kim).

**Table 1**  
Aromatic VOC and carbonyl compounds investigated in this study.

| Order | Compounds                        |           | CAS number      | Structural formula                                            | Molecular weight (g mole <sup>-1</sup> ) |
|-------|----------------------------------|-----------|-----------------|---------------------------------------------------------------|------------------------------------------|
|       | Full name                        | Acronym   |                 |                                                               |                                          |
| 1     | Benzene                          | B         | 71-43-2         | C <sub>6</sub> H <sub>6</sub>                                 | 78.1                                     |
| 2     | Toluene                          | T         | 108-88-3        | C <sub>6</sub> H <sub>5</sub> CH <sub>3</sub>                 | 92.1                                     |
| 3     | Ethyl benzene                    | E         | 100-41-4        | C <sub>6</sub> H <sub>5</sub> CH <sub>2</sub> CH <sub>3</sub> | 106                                      |
| 4     | Meta para xylene                 | MPX       | 108-38-3        | C <sub>6</sub> H <sub>4</sub> C <sub>2</sub> H <sub>6</sub>   | 106                                      |
| 5     | Styrene                          | STY       | 100-42-5        | C <sub>6</sub> H <sub>5</sub> CH=CH <sub>2</sub>              | 104                                      |
| 6     | Total Volatile Organic Compounds | TVOC      | NA <sup>a</sup> | NA                                                            | NA                                       |
| 7     | Formaldehyde                     | Form-A    | 50-00-0         | CH <sub>2</sub> O                                             | 30.0                                     |
| 8     | Acetaldehyde                     | Acet-A    | 75-07-0         | CH <sub>3</sub> CHO                                           | 44.1                                     |
| 9     | Acrolein                         | Acrolein  | 107-02-8        | CH <sub>2</sub> =CH-CHO                                       | 56.1                                     |
| 10    | Acetone                          | Acetone   | 67-64-1         | CH <sub>3</sub> COCH <sub>3</sub>                             | 58.1                                     |
| 11    | Propionaldehyde                  | Propion-A | 123-38-6        | CH <sub>3</sub> CH <sub>2</sub> CHO                           | 58.1                                     |
| 12    | Crotonaldehyde                   | Croton-A  | 123-73-9        | CH <sub>3</sub> CH=CHCHO                                      | 70.1                                     |
| 13    | Butyraldehyde                    | Butyr-A   | 123-72-8        | CH <sub>3</sub> CH <sub>2</sub> CH <sub>2</sub> CHO           | 72.1                                     |
| 14    | Benzaldehyde                     | Benz-A    | 100-52-7        | C <sub>6</sub> H <sub>5</sub> CHO                             | 106                                      |
| 15    | Valeraldehyde                    | Valer-A   | 110-62-3        | CH <sub>3</sub> (CH <sub>2</sub> ) <sub>3</sub> CHO           | 86.1                                     |

<sup>a</sup> NA; not applicable.

during the combustion of barbecue charcoal, 16 charcoal samples produced from 4 countries (4 from Korea, 7 from Indonesia, 4 from China, and 1 from Malaysia) and commonly available in South Korea were investigated. To collect each sample, an equal amount (540 g) of charcoal was placed in an old-style Korean combustor (Fig. 1). Emission gases released from charcoal combustion were collected in 10-L Tedlar bags with the aid of a lung sampler (ACEN Co. Ltd., Korea). The collection of these gas samples was made for 30 min (after passing by the early emission gases for 5 min) by slowly releasing vacuum created inside a lung sampler through a Teflon sampling line connected to the smoke stake (or chimney) (Fig. 1).



1. Body of the combustor
2. Cover of the combustion chamber
3. Water pipe
4. Combustion chamber
5. Inlet-outlet of water pipe
6. Burning charcoal
7. Chimney (or smoke stake)
8. Sampling line: connected between chimney and lung sampler
9. Inlet valve
10. Tedlar bag (10 L)
11. Lung sampler

**Fig. 1.** Schematic diagram of an old-style Korean combustor.

For the collection of carbonyls, these gas samples were passed through LpDNPH cartridges (Supelco Inc., USA) at a normal set-up value (at a fixed sampling flow rate 1 L min<sup>-1</sup>) for 5 min via a Sep-Pak ozone scrubber (Waters, US). Each cartridge was placed in the upstream to avoid artifact interferences caused by the degradation of carbonyl-hydrazones [13]. The basic physicochemical properties (e.g., structural formula, molecular weight, and CAS number) of the target compounds are briefly summarized in Table 1.

## 2.2. Analysis

Because all of our sample collection was made under laboratory conditions, the analysis of target compounds (or any additional handling of samples like cartridge treatment) was made within 24 h of sample collection. The analysis of the VOCs was conducted by combining a gas chromatography system (GC: Model 6200, Donam Instrument, Korea) equipped with a flame ionization detector (FID) and a thermal desorber (TD: Markes Ltd., UK). First, the gaseous samples in a Tedlar bag were passed through the cold trap unit of the thermal desorber (TD) at a pre-concentration of  $-10^{\circ}\text{C}$ . Then, the VOCs were thermally desorbed and transferred into the GC system and separated on a DB-VRX column (60 m length, 0.32 mm ID, 1.8  $\mu\text{m}$  film thicknesses, Agilent, USA). Finally, they were detected with an FID. To conduct VOCs analysis, the system was operated in the following sequence: (1) the samples (up to 0.3 L) were transferred into a TD unit; (2) the analytes were caught up on a cold trap at  $-10^{\circ}\text{C}$ ; (3) they were thermally desorbed at  $320^{\circ}\text{C}$  for 5 min; and (4) the thermally desorbed analytes were transferred to the FID system. The cold trap was prepared by packing Carboxen 1000 (60/80 mesh) and Carboxen 1000 (60/80 mesh) at a 1:1 mass ratio basis for the adsorbents (Supelco, US).

The GC/FID system was operated at the following temperature ( $T$ ) settings: (1)  $T$  (initial) =  $35^{\circ}\text{C}$  (for 2 min), (2)  $T$  (ramping) =  $5^{\circ}\text{C min}^{-1}$  rate, and (3)  $T$  (final) =  $175^{\circ}\text{C}$  (for 0 min). The VOCs were then separated on DB-VRX chromatographic column at a column flow rate of  $1.5\text{ mL min}^{-1}$  ( $\text{N}_2$  carrier gas). The entire running cycle for each analysis was programmed to end at 30 min intervals. The detector temperature was set as  $240^{\circ}\text{C}$ , while gas flows were maintained at  $\text{H}_2 = 30$ ,  $\text{N}_2 = 30$ , and air =  $300\text{ mL min}^{-1}$ .

As part of quality assurance, standards of BTEX prepared gravimetrically (EPA VOC Mix 2 (Supelco, USA)) were tested against gaseous standards diluted to the range from commercially purchased ones (Rigas, Korea). The results were fairly agreeable by differences in less than a few percent. In addition to the quantifica-

tion of individual VOC, 'Total Volatile Organic Compounds' (TVOCs) was also calculated as the sum of all hydrocarbon compounds (in ppbC) between C<sub>6</sub> and C<sub>16</sub> to facilitate the interpretations of VOC behavior [14]. Note that TVOCs are frequently listed in the Material Safety Data Sheet (MSDS) or technical specification information provided by a manufacturer. The addition of a new parameter was helpful, as it compromised with detailed descriptions of the nature and effects of individual constituents. The detection limits of aromatic VOCs, if expressed in terms of absolute mass, were commonly achieved at the 0.1 ng level.

For the analysis of carbonyls, each cartridge was wrapped in an aluminum foil after sampling and stored at 4 °C before injection into the HPLC. The analysis was performed by HPLC (Lab Alliance 500) equipped with a UV detector and dsCHROM software for peak integration. To initiate the analysis, the cartridges were eluted slowly with acetonitrile and filtered through 0.45 μm, 13 mm, GHP Acrodisc filters (PALL, NY, USA) into 5 mL capacity borosilicate glass volumetric flask. The eluate was manually injected into the HPLC system equipped with a 20 μL sample loop. Carbonyl-hydrazones were separated on a Hichrom 250 mm × 4.6 mm ODS (octadecyl silane), 5 μm reverse phase C<sub>18</sub> column using a mobile phase of acetonitrile + water (7:3 by volume) at a flow rate of 1.5 mL min<sup>-1</sup> and at a wavelength of 360 nm. Quantification of the carbonyls was performed against four point calibration curves drawn at 0.15, 0.3, 0.6, and 1.2 ng μL<sup>-1</sup> (at 20 μL injection volume). Liquid phase standards were prepared from the carbonyl-DNPH Mix (Supelco, USA) at a wavelength of 360 nm.

The DL values (in absolute mass) for the carbonyl species were estimated by multiplying the standard deviation (SD) values of the least detectable quantities (seven replicate analyses) by a factor of 3.14. The DL values, if expressed in terms of mixing ratio (assuming a total sampling volume of 15 L), fell in the range of 0.92 (acrolein) to 1.16 ppb (valeraldehyde).

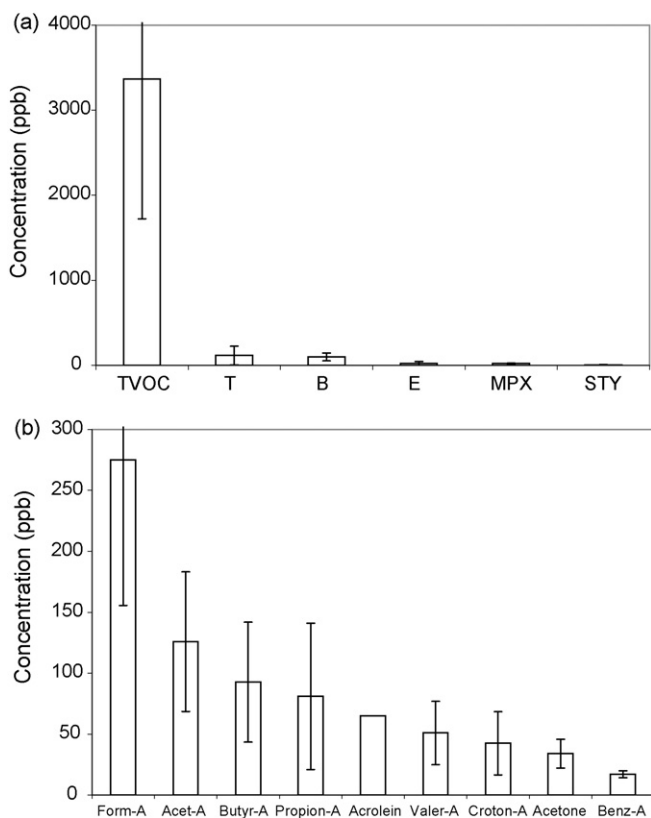


Fig. 2. Comparison of mean emission concentrations of VOC and carbonyl compounds for charcoal products investigated in this study.

Table 2  
Emission concentrations of VOC and carbonyl compounds investigated during charcoal combustion in this study (concentration in ppb).

| Order | Short name <sup>a</sup> | Country source    |      |      |           |       |      |       |      |      |          |       |      |      |      |      |      |
|-------|-------------------------|-------------------|------|------|-----------|-------|------|-------|------|------|----------|-------|------|------|------|------|------|
|       |                         | Korea             |      |      | Indonesia |       |      | China |      |      | Malaysia |       |      |      |      |      |      |
|       |                         | K1 <sup>b</sup>   | K2   | K3   | K4        | I1    | I2   | I3    | I4   | I5   | I6       | I7    | C1   | C2   | C3   | C4   | M1   |
| 1     | B                       | 18.9              | 148  | 86.6 | 137       | 168   | 19.6 | 752   | 15.6 | 4.19 | 88.8     | 17.4  | 56.7 | 8.41 | 10.6 | 23.4 | 23.7 |
| 2     | T                       | 2.87              | 7.53 | 3.56 | 1780      | 4.86  | 3.48 | 5.55  | 3.62 | 3.23 | 14.1     | 3.91  | 12.0 | 4.41 | 5.28 | 3.65 | 2.71 |
| 3     | E                       | 0.11 <sup>c</sup> | 0.11 | 0.11 | 111       | 0.739 | 0.09 | 0.11  | 0.11 | 0.11 | 0.11     | 0.089 | 0.11 | 0.11 | 1.52 | 0.11 | 0.11 |
| 4     | MPX                     | 33.4              | 1.90 | 27.4 | 43.1      | 0.04  | 0.04 | 28.3  | 0.04 | 0.04 | 13.5     | 10.2  | 6.51 | 2.40 | 20.7 | 22.8 | 37.8 |
| 5     | STY                     | 0.12              | 0.12 | 0.12 | 16.2      | 0.12  | 0.12 | 0.12  | 0.12 | 0.12 | 2.52     | 0.12  | 0.12 | 1.39 | 0.12 | 0.12 | 0.12 |
| 6     | TVOC <sup>d</sup>       | 560               | 1687 | 567  | 25,820    | 4494  | 1869 | 2142  | 1225 | 1330 | 11,230   | 587   | 1211 | 203  | 336  | 387  | 315  |
| 1     | Form-A                  | 301               | 83.4 | 117  | 72.6      | 184   | 101  | 272   | 59.8 | 45.9 | 507      | 221   | 143  | 54.3 | 2008 | 120  | 110  |
| 2     | Acet-A                  | 84.8              | 19.9 | 49.0 | 79.8      | 25.5  | 48.1 | 17.3  | 22.0 | 9.12 | 747      | 89.3  | 56.3 | 10.6 | 670  | 24.0 | 61.4 |
| 3     | Acrolein                | 0.92              | 0.92 | 0.92 | 0.92      | 0.92  | 0.92 | 0.92  | 0.92 | 0.92 | 65.0     | 0.92  | 0.92 | 0.92 | 0.92 | 0.92 | 0.92 |
| 4     | Acetone                 | 24.8              | 13.7 | 14.6 | 19.3      | 8.78  | 20.8 | 15.7  | 15.5 | 12.9 | 93.3     | 29.3  | 36.5 | 14.1 | 195  | 14.9 | 15.0 |
| 5     | Propion-A               | 1.03              | 1.03 | 1.03 | 1.03      | 1.03  | 1.03 | 1.03  | 1.03 | 1.03 | 16.2     | 22.3  | 1.03 | 1.03 | 1.03 | 1.03 | 1.03 |
| 6     | Croton-A                | 0.98              | 0.98 | 0.98 | 21.1      | 0.98  | 0.98 | 11.5  | 0.98 | 0.98 | 1.04     | 15.4  | 0.98 | 0.98 | 1.21 | 0.98 | 0.98 |
| 7     | Butyr-A                 | 1.04              | 1.04 | 1.04 | 1.04      | 1.04  | 1.04 | 1.04  | 1.04 | 1.04 | 230      | 1.04  | 1.04 | 1.04 | 97.1 | 1.04 | 1.04 |
| 8     | Benz-A                  | 0.98              | 0.98 | 0.98 | 0.98      | 0.98  | 0.98 | 0.98  | 0.98 | 0.98 | 19.9     | 0.98  | 0.98 | 0.98 | 14.1 | 0.98 | 0.98 |
| 9     | Valer-A                 | 1.16              | 1.16 | 1.16 | 1.16      | 1.16  | 1.16 | 1.16  | 1.16 | 1.16 | 76.9     | 1.16  | 1.16 | 1.16 | 24.8 | 1.16 | 1.16 |

<sup>a</sup> Refer to Table 1 for full name.

<sup>b</sup> Sample code: K, Korea; I, Indonesia; C, China; M, Malaysia.

<sup>c</sup> Underlined values represent below detection limit (BDL) values.

<sup>d</sup> TVOC in ppbC.

### 3. Results and discussion

#### 3.1. The general features of VOC emissions during combustion

In Table 2, all the measurement data collected in this study are presented as raw data to allow a direct comparison of emission patterns between different charcoal products. The emission characteristics of VOCs varied significantly with sample type, although the smoke from each sample generally contained large quantities of aromatic compounds. The mean emission concentrations of the VOCs (ppb) decreased on the order: toluene (116), benzene (98.7), ethyl benzene (22.7), m,p-xylene (20.7), and styrene (6.70) (Fig. 2a). From one sample (i.e., K4), the toluene value was exceptionally high (i.e., 1780 ppb) relative to other samples, although the second highest value was only 14.1 ppb. In light of the variability for toluene data, we simultaneously considered its median values along with the mean. Compared simultaneously by both criteria, the emissions of benzene are more prominent than toluene. The importance of benzene and toluene as major ingredients of charcoal exhaust gases can be stressed further by their presence in all 16 samples, while other VOCs were not observed as frequently as these two.

The emission concentrations of each charcoal sample were compared among countries in Table 3. Here, TVOC values for the Korean products tended to be higher than those from other countries. The TVOC results for the Korean samples recorded the highest value (7160 ppbC), while those of from Indonesia, China, and Malaysia were much smaller at 3270, 510 and 315 ppbC, respectively. This indicates that the VOC levels of Korean products can be clearly distinguished from those of the other three countries. Considerably large metal levels were also seen from Korean charcoal products investigated recently by our study group, although those samples were not identical to the ones used in this study [15]. In contrast, the emission concentrations from the Chinese and Malaysian samples were similar and showed reduced values (Fig. 3a). This suggests that they may have been produced using the materials of similar quality and manufacturing technique.

Because of lack of database on the emissions of VOC from barbecue charcoal combustion, our charcoal emission data cannot be meaningfully compared with other comparable charcoal data sets. However, as shown in Table 4a, our results can be compared with other major outdoor sources of VOCs such as fossil fuel combustion (i.e., gasoline and diesel engines), gasoline evaporation, solvent usage, industrial production, and bio-decomposition of wastes. The comparison of VOC emission concentrations among different source types indicates that the emission concentrations of charcoal samples were high relative to the numerous industrial source processes but far lower than vehicular activity or plywood combustion. Interestingly, the relative dominance of toluene over benzene was not prominent in charcoal data unlike most other source types or ambient air samples [16,17].

#### 3.2. Carbonyl emission pattern

The concentrations of all the carbonyls measured from charcoal combustion in this study are summarized in Table 2. Three carbonyls (including formaldehyde, acetaldehyde, and acetone) were found consistently above DL in every sample. Although nine other carbonyl compounds were quantified at or above DL, they were found rather inconsistently compared to the three aforementioned compounds (Table 2). As shown in Fig. 2b, formaldehyde (275 ppb) was the most abundant carbonyl compound followed by acetaldehyde (126 ppb). The concentration levels of carbonyl compounds were notably high in two (I6 and C3) relative to all other samples (Table 2). As such, the relative dominance pattern of the carbonyl compounds was considerably different from those of VOC data sets.

In Table 3, the mean concentrations of carbonyls are compared between products from different countries. According to this analysis, the mean and median emission concentrations of Chinese charcoal were generally higher than other countries. As shown in Fig. 3b, the results of Korean and Malaysian samples showed their emission concentrations in the similar magnitude but were notably lower than others. Although a number of samples taken in this study were not sufficiently large enough to derive statistically meaningful interpretations, the results can help derive a general picture of volatile pollutant emissions associated with charcoal combustion. For instance, our observation suggests that the source properties of the two chemical groups (VOCs and carbonyls) contained in charcoal samples can be distinguished from each other.

Carbonyl compounds are of environmental concern because of their environmental prevalence and their potential to cause adverse health effects [13,18]. Ambient carbonyls are directly discharged from primary sources such as exhaust from motor vehicles and incomplete combustion of hydrocarbon fuels [19]. They can also be formed from secondary source processes such as photochemical oxidation of atmospheric hydrocarbons [20]. Typical indoor carbonyl sources include (1) off gassing from building and furnishing materials and (2) emissions from certain consumer products. Combustion processes (such as tobacco smoking, cooking, and heating) are also sources of carbonyls [21].

Although carbonyl compounds are ubiquitous in the ambient atmosphere and indoors, relatively little is known about their emissions from barbecue charcoal combustion. As a means to evaluate their emission concentrations from our experimental data, the charcoal results in this study were compared with other source types reported previously (Table 4b). The carbonyl concentrations were high in these source processes as (1) industrial and (2) traffic related activities (the bus station and restaurant). The road sources may have been due to the diesel exhaust emissions and incomplete combustion of hydrocarbon fuels. In Table 4b, three carbonyls (formaldehyde, acetaldehyde, and acetone) from each type of sources accounted for 80–85% of total carbonyl emissions. This is also comparable to the data sets derived from charcoal combustion in this study. Although the estimates of emission factors from charcoal combustion were comparably low with respect to other source types, the results of our study suggest that charcoal combustion can be fairly strong, depending on the charcoal product.

#### 3.3. Factors controlling VOC and carbonyl levels from charcoal

The results of correlation analysis between all target pollutants in this study are presented in Table 5. Because the number of quantified data differs greatly among different compounds, this analysis was confined to the pollutants measured 10 times or above ( $n \geq 10$ ). According to this analysis, the significantly correlated cases were seen occasionally across all matching pairs (4 out of 21 cases). However, carbonyl compounds (form-A, acet-A, and acetone) are tightly correlated to each other in all matching pairs, while this was not the case for the VOCs. Hence, the results of correlation analysis suggest that the relative behavior of different pollutants emanating from barbecue charcoal combustion can be distinguished between two chemical groups of VOCs and carbonyl compounds.

Charcoal is a complex organic substance containing moisture, wood ash, carbon, hydrogen, oxygen, and a variety of volatiles [1]. The raw material, particle size, and the process involved in charcoal production may play an important role in the quality of the charcoal. As a result, the nature of wood can seriously affect the concentration levels of pollutants (e.g., polycyclic aromatic hydrocarbon) released in the form of the generated smoke [22–24]. Charcoal for barbecue use typically contains 20–30% of volatiles, whereas metallurgical charcoals often contain 10–15% or even less volatile matter [25]. Comparative analysis of our data suggests that

**Table 3**  
Comparison of emission concentration levels (ppb) of VOC and carbonyls in different countries<sup>a</sup>.

| Country source |          | VOC    |      |      |      |      |                   | Carbonyl compounds |        |          |         |           |          |         |        |         |      |
|----------------|----------|--------|------|------|------|------|-------------------|--------------------|--------|----------|---------|-----------|----------|---------|--------|---------|------|
|                |          | B      | T    | E    | MPX  | STY  | TVOC <sup>b</sup> | Form-A             | Acet-A | Acrolein | Acetone | Propion-A | Croton-A | Butyr-A | Benz-A | Valer-A |      |
| Korea          | Mean     | 98.6   | 448  | 111  | 26.5 | 16.2 | 7160              | 144                | 58.3   | –        | 18.1    | –         | 21.1     | –       | –      | –       |      |
|                | SD       | 58.9   | 888  | –    | 17.6 | –    | 12450             | 107                | 30.1   | –        | 5.10    | –         | –        | –       | –      | –       |      |
|                | Median   | 111    | 5.55 | –    | 30.4 | –    | 1127              | 100                | 64.4   | –        | 16.9    | –         | –        | –       | –      | –       |      |
|                | Min      | 18.9   | 2.87 | –    | 1.9  | –    | 560               | 72.6               | 19.9   | –        | 13.7    | –         | –        | –       | –      | –       |      |
|                | Max      | 148    | 1780 | –    | 43.1 | –    | 25820             | 301                | 84.9   | –        | 24.8    | –         | –        | –       | –      | –       |      |
|                | n        | 4      | 4    | 1    | 4    | 1    | 4                 | 4                  | 4      | –        | –       | –         | –        | –       | –      | –       |      |
|                | Mean     | 152    | 5.54 | 0.31 | 17.3 | 2.52 | 3270              | 199                | 137    | 65.0     | 28.1    | 113       | 13.9     | 123     | 19.9   | 76.9    |      |
|                | SD       | 271    | 3.87 | 0.38 | 9.64 | –    | 3724              | 160                | 270    | –        | 29.5    | 128       | 3.32     | 152     | –      | –       |      |
| Indonesia      | Median   | 19.6   | 3.91 | 0.09 | 13.5 | –    | 1869              | 184                | 25.5   | –        | 15.7    | 113       | 13.9     | 123     | –      | –       |      |
|                | Min      | 4.19   | 3.23 | 0.09 | 10.2 | –    | 587               | 45.9               | 9.12   | –        | 8.78    | 22.3      | 11.5     | 15.4    | –      | –       |      |
|                | Max      | 752    | 14.1 | 0.74 | 28.3 | –    | 11230             | 507                | 747    | –        | 93.3    | 203       | 16.2     | 230     | –      | –       |      |
|                | n        | 7      | 7    | 3    | 3    | 1    | 7                 | 7                  | 7      | 1        | 7       | 2         | 2        | 2       | 1      | 1       |      |
|                | Mean     | 25     | 6.34 | 1.52 | 13.1 | 1.39 | 510               | 581                | 190    | –        | 65.1    | –         | 121      | 62.5    | 14.1   | 24.8    |      |
|                | SD       | 22.3   | 3.83 | –    | 10.2 | –    | 471               | 951                | 320    | –        | 87.2    | –         | –        | 49.0    | –      | –       |      |
|                | Median   | 17     | 4.85 | –    | 13.6 | –    | 25820             | 132                | 40.2   | –        | 25.7    | –         | –        | 62.5    | –      | –       |      |
|                | Min      | 8.41   | 3.65 | –    | 2.4  | –    | 203               | 54.3               | 10.6   | –        | 14.1    | –         | –        | 27.8    | –      | –       |      |
| China          | Max      | 56.7   | 12   | –    | 22.8 | –    | 1211              | 2008               | 670    | –        | 195     | –         | –        | 97.1    | –      | –       |      |
|                | n        | 4      | 4    | –    | 4    | 1    | 4                 | 4                  | 4      | –        | 4       | –         | 1        | 2       | 1      | 1       |      |
|                | Value    | 23.7   | 2.71 | –    | 37.8 | –    | 315               | 110                | 61.4   | –        | 15      | 18        | –        | –       | –      | –       |      |
|                | n        | 1      | 1    | –    | 1    | –    | 1                 | 1                  | 1      | –        | 1       | 1         | –        | –       | –      | –       |      |
|                | Malaysia | Mean   | 98.7 | 116  | 22.7 | 20.7 | 6.70              | 3367               | 275    | 126      | 65.0    | 34.0      | 81.0     | 42.5    | 92.6   | 17.0    | 50.9 |
|                |          | SD     | 183  | 444  | 49.4 | 13.9 | 8024              | 6573               | 477    | 229      | 0       | 47.3      | 106      | 52.5    | 98.4   | 4.10    | 36.8 |
|                |          | Median | 23.6 | 4.16 | 0.74 | 21.8 | 2.52              | 1218               | 119    | 48.6     | 65.0    | 15.6      | 22.3     | 18.7    | 62.5   | 17.0    | 50.9 |
|                |          | Min    | 4.19 | 2.71 | 0.09 | 1.90 | 1.39              | 203                | 45.9   | 9.12     | 65.0    | 8.78      | 18.0     | 11.5    | 15.4   | 14.1    | 24.8 |
| Max            |          | 752    | 1780 | 111  | 43.1 | 16.2 | 25820             | 2008               | 747    | 65.0     | 195     | 203       | 121      | 230     | 19.9   | 76.9    |      |
| n              |          | 16     | 16   | 5    | 12   | 3    | 16                | 16                 | 16     | 1        | 16      | 3         | 4        | 4       | 2      | 2       |      |
| All            |          | Mean   | 98.7 | 116  | 22.7 | 20.7 | 6.70              | 3367               | 275    | 126      | 65.0    | 34.0      | 81.0     | 42.5    | 92.6   | 17.0    | 50.9 |
|                |          | SD     | 183  | 444  | 49.4 | 13.9 | 8024              | 6573               | 477    | 229      | 0       | 47.3      | 106      | 52.5    | 98.4   | 4.10    | 36.8 |
|                | Median   | 23.6   | 4.16 | 0.74 | 21.8 | 2.52 | 1218              | 119                | 48.6   | 65.0     | 15.6    | 22.3      | 18.7     | 62.5    | 17.0   | 50.9    |      |
|                | Min      | 4.19   | 2.71 | 0.09 | 1.90 | 1.39 | 203               | 45.9               | 9.12   | 65.0     | 8.78    | 18.0      | 11.5     | 15.4    | 14.1   | 24.8    |      |
|                | Max      | 752    | 1780 | 111  | 43.1 | 16.2 | 25820             | 2008               | 747    | 65.0     | 195     | 203       | 121      | 230     | 19.9   | 76.9    |      |
|                | n        | 16     | 16   | 5    | 12   | 3    | 16                | 16                 | 16     | 1        | 16      | 3         | 4        | 4       | 2      | 2       |      |

<sup>a</sup> BDL values shown in Table 2 are excluded for the derivation of each statistical parameter for this comparison.

<sup>b</sup> TVOC in ppbC.

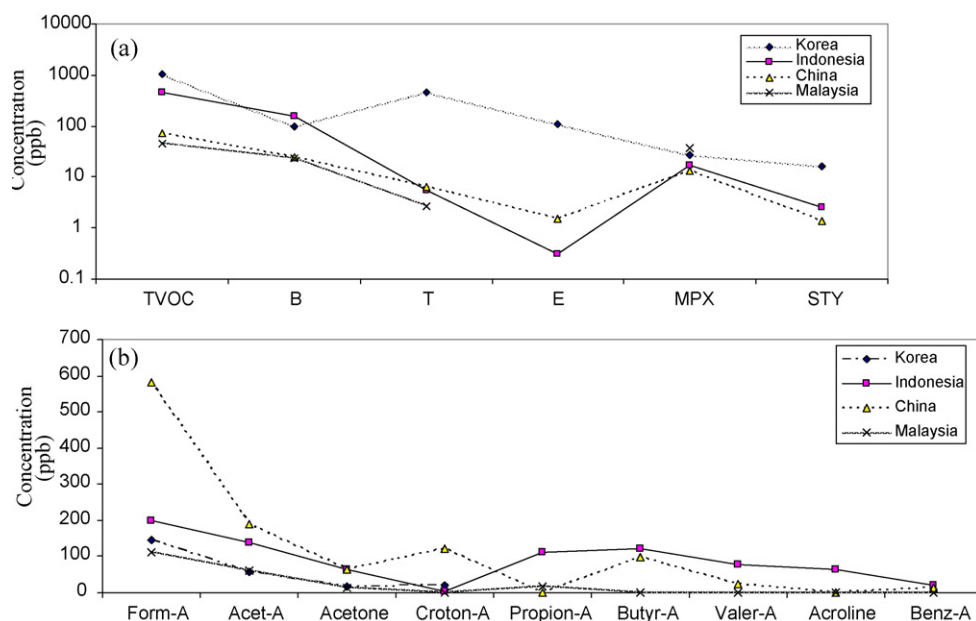


Fig. 3. Comparison of mean emission concentrations of airborne pollutants released from charcoal products of four countries: (a) VOC and (b) carbonyl compounds.

**Table 4**  
Comparison of VOC and carbonyl emissions from different sources<sup>a</sup>.

| Order                | VOC source                                     | Concentration (ppb) |         |          |         |           |          | Data source |        |            |         |             |
|----------------------|------------------------------------------------|---------------------|---------|----------|---------|-----------|----------|-------------|--------|------------|---------|-------------|
|                      |                                                | B                   | T       | E        | MPX     | STY       | TVOC     |             |        |            |         |             |
| <b>(a) VOC</b>       |                                                |                     |         |          |         |           |          |             |        |            |         |             |
| 1                    | Food and beverage industry                     | 1.21                | 35.8    | 2.08     | 2.55    | 1.44      | 1,273    | [37]        |        |            |         |             |
| 2                    | Textiles industry                              | 1.53                | 45.7    | 11.6     | 26.5    | 14.5      | 8,120    | [37]        |        |            |         |             |
| 3                    | Leather industry                               | 1.3                 | 97.5    | 21.3     | 11.3    | 6.87      | 9,412    | [37]        |        |            |         |             |
| 4                    | Pulp, paper industry                           | 4.32                | 2,466   | 12       | 14.3    | 10.4      | 21,807   | [37]        |        |            |         |             |
| 5                    | Compound and chemical product industry         | 21.1                | 277     | 201      | 112     | 61.2      | 11,275   | [37]        |        |            |         |             |
| 6                    | Sewage, waste treatment, and cleaning industry | 34.8                | 367     | 32.5     | 26.4    | 16        | 9,448    | [37]        |        |            |         |             |
| 7                    | Petrol pump                                    | 471                 | –       | –        | –       | –         | –        | [38]        |        |            |         |             |
| 8                    | Motor vehicle<br>(At chassis dynamometer)      | 26,870              | 103,890 | –        | –       | –         | –        | [39]        |        |            |         |             |
|                      |                                                | 78,270              | 92,920  | –        | 50,750  | –         | –        | [40]        |        |            |         |             |
| 9                    | Motor cycle                                    | 34,340              | –       | –        | –       | –         | –        | [41]        |        |            |         |             |
| 10                   | Plywood                                        | 7,000               | 3,000   | –        | –       | –         | –        | [42]        |        |            |         |             |
| 11                   | Charcoal                                       | 98.7                | 116     | 22.7     | 20.7    | 6.70      | 3,367    | This study  |        |            |         |             |
| Order                | Carbonyl source                                | Concentration (ppb) |         |          |         |           |          |             |        |            |         | Data source |
|                      |                                                | Form-A              | Acet-A  | Acrolein | Acetone | Propion-A | Croton-A | Butyr-A     | Benz-A | Isovaler-A | Valer-A |             |
| <b>(b) Carbonyls</b> |                                                |                     |         |          |         |           |          |             |        |            |         |             |
| 1                    | Industrial area                                | 323                 | 299     | 1.69     | 297     | 34.4      | 8.66     | 186         | 24.5   | 11.9       | 3.31    | [19]        |
| 2                    | Bus station                                    | 54.6                | 12.8    | 0.645    | 6.57    | 1.7       | 0.443    | 1.14        | 0.399  | 0.173      | 0.394   | [43]        |
| 3                    | Highway tunnel                                 | 24.9                | 5.1     | 0.218    | 1.5     | 0.357     | 1.25     | –           | 0.437  | 0.426      | 0.142   | [44]        |
| 4                    | Restaurant                                     | 54.5                | 47.2    | –        | 22.1    | 5.02      | 1.1      | 4.11        | 0.438  | 0.320      | 4.98    | [43]        |
| 5                    | Charcoal combustion                            | 275                 | 126     | 13.7     | 34      | 35.3      | 11.3     | 49.7        | 4.76   | 1.18       | 50.7    | This study  |

<sup>a</sup> 1–6 industrial sector, 7–9 vehicular activity, 10–11 combustion.

**Table 5**  
Results of correlation analysis between VOC and carbonyl components.

|         | B      | T       | MPX    | TVOC   | Form-A  | Acet-A  | Acetone |
|---------|--------|---------|--------|--------|---------|---------|---------|
| B       | 1      |         |        |        |         |         |         |
| T       | 0.057  | 1       |        |        |         |         |         |
| MPX     | 0.176  | 0.506   | 1      |        |         |         |         |
| TVOC    | 0.114  | 0.913** | 0.409  | 1      |         |         |         |
| Form-A  | –0.056 | –0.112  | 0.001  | –0.087 | 1       |         |         |
| Acet-A  | –0.128 | –0.049  | –0.080 | 0.196  | 0.796** | 1       |         |
| Acetone | –0.151 | –0.081  | –0.091 | 0.010  | 0.963** | 0.887** | 1       |

The value of E, STY, acrolein, propion-A, croton-A, butyr-A, benz-A, and valer-A are not considered for the correlation analysis as most of the values are below detection limit.

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

**Table 6**  
Permissible exposure limits (PEL) of short term exposure levels (STEL) for carbonyl compounds<sup>a</sup>.

| Substance | STEL (ppm)      | Mean values (ppm) | Maximum emission from the sample (ppm) | Sample code |
|-----------|-----------------|-------------------|----------------------------------------|-------------|
| Form-A    | 2               | 0.275             | 2.008                                  | C3          |
| Acet-A    | 150             | 0.126             | 0.747                                  | I6          |
| Acrolein  | 0.3             | 0.065             | 0.065                                  | I6          |
| Acetone   | 1000            | 0.034             | 0.195                                  | C3          |
| Propion-A | NA <sup>b</sup> | 0.081             | 0.203                                  | I6          |
| Croton-A  | 2               | 0.043             | 0.121                                  | C3          |
| Butyr-A   | 2               | 0.093             | 0.230                                  | I6          |
| Benz-A    | NA              | 0.017             | 0.019                                  | I6          |
| Valer-A   | NA              | 0.051             | 0.077                                  | I6          |

<sup>a</sup> Source: [31,32].

<sup>b</sup> Not available.

the emission patterns of VOCs from barbecue charcoal combustion were affected by their manufacturing process and the raw materials used in production. Manufacturers use hardwoods such as beech, birch, hard maple, hickory, and oak as well as softwoods including long leaf and slash pine, nutshells, and fruit pits. They also use vegetable wastes, paper mill residues, sawdust and wood scraps from lumber mills, and old household furniture combined with binding agents for holding their shapes [26]. However, many commercially available brands are also known to contain potentially harmful ingredients, including coal dust (as a heat source), sodium nitrate (to aid ignition), and VOC-forming lighter fluid for quick-light brands [6]. Although we were not able to analyze the major elemental composition of our charcoal samples, the chemical properties of typical barbecue charcoal were typically fixed with carbon 50%, volatile 30%, ammonia 20%, moisture 5% and sulfur 0.8% [26].

During the production of charcoal, volatile matter (other than water) in charcoal comprised liquid and tarry residues not fully driven from the process of carbonization [25]. If carbonization is prolonged at high temperature, the content of the volatiles is low. If the carbonization temperature is low and time in the kiln is short, then the volatile matter content increases. At low temperatures (e.g., 300 °C), a charcoal yield of nearly 50% is possible. As volatiles are lower at carbonization temperatures of 500–600 °C, retort yields of 30% are typical [25]. At very high temperatures (around 1000 °C), the volatile content is almost zero. Hence, yields can fall nearly 25% [27]. Charcoal can reabsorb tars and pyrolytic acids from rain wash in pit burning and similar processes. Thus, the charcoal may maintain high volatile matter contents because of this factor. Highly volatile charcoal is easy to ignite but accompanies a smoke flame. In contrast, low volatile charcoal tends to burn cleanly despite ignition difficulty [27].

#### 3.4. Potential health risk of charcoal combustion

The presence of VOCs in ambient and indoor air is widely recognized as serious human health risks. VOCs include a variety of chemicals, of which some may have short and long-term effects. As barbecue charcoal is burnt indoors in some parts of the world (e.g., in many restaurants in Korea), their indoor concentrations can be significantly higher under certain circumstances.

The potential health effects of organic chemicals are influenced by many factors including level of exposure and length of duration [7]. Eye and respiratory tract irritation, headaches, dizziness, visual disorders, and memory impairment are among the immediate symptoms that some people can experience following exposure [7]. At present, not much is known about the health effects associated with barbecue charcoal combustion. Several organic compounds released during combustion can cause cancer in animals. Some have been demonstrated, while others are suspected to be carcinogenic [6]. There is an indication that producers may use improper raw

materials for the production of charcoal such as hardwood lumber from furniture or flooring materials containing various pollutants. As used furniture or flooring is coated with paint, they can also act as a source of VOCs, when combusted.

As air is the most effective route of VOC transmission relative to others (food, beverages, or drinking water), an intake of higher levels of VOCs generally occurs through air [28]. Benzene is readily absorbed into the body, when breathed into the lungs; about half of benzene is retained [29]. As benzene is more soluble in fat than in water, it tends to be distributed in the body in fatty tissues including the brain and the bone marrow where cells are made [29]. Accurate information on benzene in air is highly valuable. According to the Agency for Toxic Substances and Disease Registry (ATSDR) and Occupational Safety and Health Administration (OSHA) in the USA, the permissible exposure level (PEL) of benzene and toluene is 1000 and 200,000 parts per billion (ppb), respectively [30–32]. In this study, the emission concentrations for benzene and toluene fell below the PEL.

Table 6 lists permissible exposure limits (PELs) of carbonyls set by the OSHA such as short term exposure level (STEL) (i.e., maximum exposure allowed during a 15 min period) [31,32]. For formaldehyde measured in one sample (i.e., C3), the value slightly exceeded the PEL. Except this, emission concentrations of all other carbonyls were below the PEL. Airborne concentrations of formaldehyde above 100 ppb cause irritation of the eyes, nose, and throat [33]. The severity of this irritation increases with concentration increase. The upper respiratory tract irritation can potentially exacerbate asthma symptoms and other respiratory illnesses [33–35]. Dermal contact with skin can also cause various reactions including sensitization [33]. However, acute and chronic health effects of formaldehyde vary in individuals. The typical threshold for the development of acute symptoms from inhaled formaldehyde is 800 ppb [33]. However, sensitive individuals have reported symptoms at formaldehyde levels around 100 ppb [33,36]. The emission concentration of formaldehyde above 100 ppb was recognized in 11 out of 16 samples.

Acetaldehyde is a substance which may reasonably be considered carcinogenic, according to the seventh annual report on carcinogens, National Toxicology Program, U.S. Department of Health and Human Services (1994). It is also classified in the EPA's toxic release inventory (TRI) as a known or suspected carcinogen [7]. Acetaldehyde, when ingested or inhaled can irritate the eyes, nose, and throat. It can also cause conjunctivitis, coughing, central nervous system depression, eye and skin burns, dermatitis, and delayed pulmonary edema. Human exposure to other carbonyl compounds can also cause irritation of the eyes and respiratory system, mood swings, nausea, and drowsiness [28].

#### 4. Conclusion

In this work, the concentrations of VOC and carbonyl compounds emitted during the combustion of barbecue charcoals were

investigated. The total emission concentrations of individual VOCs and carbonyls were the highest in Korean and Chinese products, respectively. Benzene and toluene were the most abundant VOCs emitted from the samples. In the case of carbonyl compounds, formaldehyde was the most abundant carbonyl along with acetaldehyde. The emission concentrations levels of the VOCs and carbonyl compounds were below the PEL set by OSHA with a single exception. To date, charcoal is not listed as a hazardous product for control except in a few countries (e.g., Canada). However, some cautions are necessary, as the emission concentrations of certain pollutants were significantly high in certain samples. Concerned authorities must create a database to place effective control on the quality of charcoal production and use. They must also encourage people to practice green barbecuing procedures to help sustain forests. In view of extensive charcoal consumption worldwide, it is important to accurately characterize its potential role in environment pollution and its impact on human health.

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