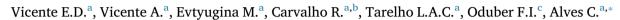
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Research article

Particulate and gaseous emissions from charcoal combustion in barbecue grills



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ABSTRACT

The use of charcoal for cooking and heating can be a major source of air pollution and lead to a wide range of health outcomes. The aim of this study was to experimentally quantify and characterise the gaseous and particulate matter (PM_{2.5}) emissions from charcoal combustion in a typical brick barbecue grill. The gaseous emission factors were $219 \pm 44.8 \, g \, kg^{-1}$ for carbon monoxide (CO), $3.01 \pm 0.698 \, g \, kg^{-1}$ for nitrogen oxides (NO_x expressed as NO₂), and $4.33 \pm 1.53 \, g C \, kg^{-1}$ for total organic carbon (TOC). Particle emissions (7.38 $\pm 0.353 \, g \, kg^{-1}$ of dry charcoal burned) were of the same order of magnitude as those from traditional residential wood burning appliances. About 50% of the PM_{2.5} emitted had a carbonaceous nature while water soluble ions accounted, on average, for 17% of the particulate mass. Alkanes (C₁₁-C₁₆ and C₂₃), hopanes, steranes and alkyl-PAHs accounted for small mass fractions of PM_{2.5}. Phenolic compounds and saccharides represented the major particle-bond organic constituents. The high proportion of either resin acids or syringyl and vanillyl compounds is consistent with emissions from charred coniferous wood. The ratios between anhydrosugars for charcoal are much lower than the values reported for lignite combustion, but overlap those from other biomass burning sources.

1. Introduction

In 2014, about 53 million tonnes of wood charcoal were produced worldwide [1]. Charcoal is a product of thermochemical conversion of biomass by pyrolysis and has advantages as fuel when compared to the original feedstock (biomass) such as the higher heating value and easy storage [2,3]. It has also an advantage in comparison with other renewable fuels: the cheaper production. In contrast with mineral coals, charcoal has relatively low content of sulphur or mercury, which is a benefit from the emissions point of view [2]. In the developing countries, charcoal is still an important cooking fuel [4]. Despite the spatial and temporal changes in fuel consumption patterns, charcoal remains a popular cooking fuel in the developed world since it produces food with unique flavour and texture. In fact, charcoal-grilling is extensively used by households and restaurants [3,5]. Johnson [3] estimated the charcoal grilling footprint to be 6.7 kg CO2e per grill session which is, according to the author, similar to the carbon footprint obtained for driving an average car for approximately 35 km. Charcoal production is the dominant process in the carbon footprint (45%), as well as charcoal combustion (40%). Compared to liquefied petroleum gas (LPG) grilling,

charcoal has a carbon footprint 2.9 times higher [3].

Charcoal production in Africa accounted for 61% of global production in 2014. In the Latin America and Caribbean, charcoal production grew from 2010 to 2014, reaching 10 million tonnes [1]. Several studies focused on the emissions from charcoal production have been performed [5,7-10]. In 2009, the greenhouse gas emissions arising from charcoal production in tropical ecosystems were estimated to be around 71.2 million tonnes of CO₂ and 1.3 million tonnes of CH₄ [6]. Charcoal is produced from wood pyrolysis in kilns and the process may take up to a few weeks [2]. The efficiency of traditional methods for charcoal production is about 10% to 22%. Retort kilns have been claimed to increase energy efficiency and to decrease air pollution effects [5]. Sparrevik et al. [7] confirmed that retort kilns lead to significantly lower emissions of incomplete combustion products. However, the efficiency from these kilns is lowered due to the wood consumption for start-up. In addition to the production process, charcoal combustion is a source of airborne pollution. Intense outdoor barbecue cooking during a big festival event was reported to increase the ambient PM_{10} levels by approximately 5% [11]. Charcoal burning leads to the emission of a wide range of pollutants. Alves et al. [12]

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collected $PM_{2.5}$ samples from the exhaust stacks of restaurants with different cooking styles including a charcoal grilled chicken restaurant. The $PM_{2.5}$ concentrations reported by the authors were in the range of 26 to 127 mg m⁻³. For the same sites, the VOC concentrations were also assessed [13]. The authors observed that chlorinated VOCs were only detected in samples from the charcoal grilled chicken restaurant. Benzene was the compound with the highest emission rate (201 kg year⁻¹).

In indoor environments, such as restaurants, personal exposure to toxic pollutants from charcoal burning can be significantly high, posing a health risk to the customers and employees. Lee et al. [14] assessed the indoor air quality at four different types of restaurants. The authors found the highest CO $(15,100 \,\mu g \,m^{-3})$ and particulate concentrations (1442 and 1167 μ g m⁻³ PM₁₀ and PM_{2.5}, respectively) at the barbecue style restaurant. High PM_{10} (15,074 µg m⁻³) and $PM_{2.5}$ $(13,700 \,\mu g \,m^{-3})$ concentrations were also found in a Korean-style barbecue restaurant in Seoul [15]. Moreover, barbecue cooking can generate considerable amounts of benzene, toluene, methylene chloride and chloroform [14]. Chinese traditional charbroiling was found to be a source of particle-bound polycyclic aromatic hydrocarbons (PAHs) and fatty acids indoors. The latter accounted for over 90% of all identified organic compounds. However, PAHs had a major contribution to the particulate mass [16]. Taner et al. [17] carried out a study at 14 nonsmoking restaurants in Turkey. The authors concluded that fine particles associated with charcoal cooking represented a significant source of indoor pollution. Charcoal combustion played an important role in the PM trace element content and the most abundant elements identified were As, Cr, Se, V, and Zn.

Pollutant emissions from charcoal burning are associated with the raw materials nature and the charcoal production process [18-21]. Kabir et al. [22] investigated trace metal emissions from eleven types of charcoal available in the Korean market (Korean and imported products). The combustion tests were carried out in an old type Korean burner. Although the trace metal concentrations varied among different charcoal types, Fe and Zn were consistently the most abundant metals. In an earlier study, Kabir et al. [19] reported aromatic volatile organic compounds and carbonyl emissions from the same charcoal types. Benzene and toluene were the most abundant VOCs, while formaldehyde and acetaldehyde were the main carbonyls. The predominance of benzene over other aromatic compounds emitted from glowing charcoal was also reported by Olsson and Petersson [20]. Charcoal burning is also a source of offensive odorants [21]. Barbecue charcoal combustion can be a significant source of trace metal emissions to the atmosphere. Susaya et al. [23] reported concentrations of Cd, Co and Ni exceeding the inhalation minimum risk levels of the United States Agency for Toxic Substances and Disease Registry for chronic duration exposure. In their study, eleven types of charcoal were burned in a combustion device built in a traditional Korean style with stainless-steel vent line. The major PM-bound metal elements were Zn and Pb. Although these and other studies provide remarkable data on emissions from charcoal burning, only a few aimed at characterising the PM chemical composition, and these evaluations were mainly focused on the elemental content. Particles released during charcoal burning contain a broad range of chemical species, ranging from elemental to organic and inorganic compounds. Very little is known about the particulate organic speciation. Due to their carcinogenic potential, only PAHs have been addressed in a few studies [11,24,25]. In most researches, only gaseous and particulate matter concentrations were measured, not encompassing a more exhaustive characterisation of the PM samples. Furthermore, the majority of the studies on this topic have been focused on particular fuels from particular regions. A rather extensive amount of work has been performed to characterise the emissions from charcoal of the Asian market (e.g., [18,19,21-23,25]), and, to a lesser extent, few studies focused on charcoal fuels from Poland [20], Sweden [20,24] and United States [21,26]. The raw materials used in charcoal production, as well as the production process conditions, are very important regarding the final properties of the fuel [2].

These, in turn, are key aspects concerning the emissions generated by charcoal combustion (e.g., [18]). For these reasons, it is advantageous to obtain source-specific emission profiles taking into account the fuel specificities.

The aim of this study was to experimentally quantify and characterise the particulate and gaseous emissions from charcoal combustion in a typical Portuguese barbecue grill. In doing so, the data presented in the current study will improve emission inventories, which are indispensable to establish environmental measures to prevent air pollution. Moreover, new databases of speciated emission profiles will contribute to more accurate source apportionment results when applying receptor models.

2. Methodology

2.1. Combustion infrastructure, fuel and experimental procedure

The experimental setup of the present study replicates the Portuguese charcoal barbecue grills, which have side and back walls made of refractory brick and a chimney hood. The experimental setup included a grate where the charcoal was burned and which was placed over a weight sensor (DSEUROPE Model 535QD-A5), allowing the continuous monitoring of the fuel mass during the combustion cycles. The combustion flue gas flow rate across the chimney was calculated by monitoring the gas velocity with a Pitot tube connected to a differential pressure transmitter (JUMO, Type 404304). A detailed description of the experimental setup can be found elsewhere [27].

The charcoal used in the combustion experiments was purchased from a local supplier. The fuel properties were determined according to international CEN/TS standards and included moisture (CEN/TS 14774), ash content (CEN/TS 14775), volatile matter (CEN/TS 15148), C, H, N, S (CEN/TS 15104) and calorific value (CEN/TS 14918) (Table 1). In order to replicate the householder's practices, the tests were initiated by lightning small pieces of wood on the top of the batch of charcoal. A total of four cold start tests, initiated with the combustion appliance at ambient temperature, were conducted. The experiments were made with batches of around 1 kg of charcoal fuel and lasted approximately 1.5 to 2 h. The combustion temperature was monitored using K-type thermocouples at three locations: under the fixed bed of fuel at the grate, at the central region of the combustion chamber (0.25 m above the fixed bed of fuel) and at the chimney exit (2.6 m above the fixed bed of fuel).

2.2. Gas sampling and measurement techniques

The combustion flue gas was sampled at the chimney through a heated (at 180 $^{\circ}$ C) sampling line, which conducted the gas to an online

Table 1

Ultimate and proximate analysis of the charcoal used as fuel in the combustion experiments.

Proximate analysis (wt%, as received)	
Moisture	4.06
Ash	11.0
Volatile matter	4.36
Fixed carbon (by difference)	84.7
Ultimate analysis (wt%, dry basis)	
С	79.6
Н	2.5
Ν	1.03
S	< 0.01
O (by difference)	5.44
Lower heating value (MJ kg ⁻¹ , dry basis)	29.3

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