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COMPARISON OF COMBUSTION POLLUTANTS FROM CHARANIA BRIQUETTES, CONSUMER BARBECUE BRIQUETTES, PAKISTANI MINERAL DEVELOPMENT CORPORATION BRIQUETTES, AND PAKISTANI WOOD CHARCOAL

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Health and Safety Research Division

Comparison of Combustion Pollutants From Charania Briquettes, Consumer Barbecue Briquettes, Pakistani Mineral Development Corporation Briquettes, and Pakistani Wood Charcoal

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ABSTRACT

This report presents the results of an experimental comparison of combustion pollutants from four fuel sources: Charania briquettes, a new fuel source being evaluated for use in Pakistan; barbecue briquettes commercially available in the United States; Pakistani natural wood charcoal; and Pakistani Mineral Development Corporation briquettes (PMDC). The main objective of this study was to determine the relative emissions of carbon monoxide (CO), oxides of nitrogen (NO_x), sulfur dioxide (SO_2), photoionizable organic compounds (POC) and respirable suspended particulates (RSP) from the combustion of Charania briquettes compared to more traditional fuels.

The samples (50 g) were combusted within a room size chamber (17 m^3) at an air exchange rate of 0.78 h^{-1} with an open barbecue grill. Sampling of the air within the chamber was performed via sample lines placed approximately 1.5 m from the floor. Three fans were used within the chamber to ensure good mixing of the combustion gases.

The results obtained in this combustion study indicate that the Charania briquettes are somewhat higher in CO, SO_2, and POC than the natural wood charcoal, but are similar in RSP and lower in NO_x concentrations. In all cases, the peak CO concentrations (~350 mg/m^3) generated by these 50 g samples in the room-size test chamber are high compared to various standards (e.g., occupational, ambient air).

1. INTRODUCTION

A significant environmental impact in many countries is caused by extensive deforestation to meet heating and cooking fuel requirements. One alternative that should improve the situation is to use coal-based or

biomass fuels as substitutes for wood charcoal. The effectiveness of these fuels must be evaluated relative to the current practice of using wood charcoal. Several factors such as availability, suitability, and economic feasibility must be evaluated. One of these important factors is the production of pollutants due to combustion of these alternative fuels. This report will discuss the experimental procedures and the results obtained from a comparison of pollutant emissions from four fuel sources. They are: (1) commercial U.S. charcoal, (2) Pakistan natural wood charcoal, (3) Charania briquettes, and (4) Pakistan Mineral Development Corporation (PMDC) briquettes. Specific pollutant emissions measured were: carbon monoxide (CO), nitrogen oxides (NOx), sulfur dioxide (SO_2), photoionizable organic compounds (POC), and respirable suspended particulates (RSP).

2. EXPERIMENTAL EQUIPMENT

2.1 ENVIRONMENTAL CHAMBER

The chamber used in this study was a 17 m^3 Hotpack Environmental Room equipped with two water cooled radiators to ensure proper temperature control and three 20-in. fans to ensure good mixing within the chamber. In addition, the chamber was equipped with three Matheson Mass Flow Controllers (model 8260), each delivering 60 L/min of air. This flow enabled the chamber to obtain a stable air exchange rate of 0.78 h^{-1}.

2.2 INSTRUMENTATION

The instrumentation used in the study was as follows:

CO: Interscan CO Monitor Model 4124

NO_2: Monitor Labs Nitrogen Oxides Analyzer

SO_2: Thermo Electron Pulsed Fluorescent SO_2 Analyzer Series 43

POC: HNU Photo-Ionizer Model PI 101 equipped with a 10.2-eV lamp

Temp/RH: Campbell 207 Temperature/Relative Humidity Probe

RSP: Harvard Respirable Particulate Sampler (samples particulates <2.5 μm)

Data Acquisition: Campbell 21x Micrologger

Data Analysis: IBM PC

Software: Lotus 1-2-3

2.2 COMBUSTION VESSEL

The samples were combusted within an open Chimney Quick Charcoal Lighter apparatus. The apparatus was then placed on the bottom grill of a Sunbeam Patio Master Barbecue Grill.

2.3 SAMPLES

Three of the four samples tested were provided by researchers in the ORNL Energy Division; the fourth one, consumer barbecue briquettes, was obtained from a local retail store. The fuels tested were: (1) Pakistani wood charcoal, (2) Charania briquettes (an alternative fuel being developed in Pakistan), (3) PMDC, briquettes, a fuel being developed with support from the U.S. Agency for International Development, and (4) commercial U.S. barbecue briquettes. The known physical and chemical properties of the samples (as provided by Energy Division) are listed in Tables I and II.

Table I. Physical properties of the samples tested

Sample	Moisture (%)	Fixed carbon (%)	Density* (g/cc)
Pakistani			0.5
Charania	2.7	47.84	1.2
Barbecue			1.2
PMDC	3.3	42.52	4.1

*Measured by HASRD staff.

Table II. Elemental analysis of Charania and PMDC briquettes

Element	Charania (wt %)	PMDC (wt %)
Hydrogen	3.2	3.94
Carbon	57.76	45.52
Nitrogen	1.0	0.89
Sulfur	0.75	7.18
Oxygen	18.51	10.25
Ash	18.75	35.19
Volatile matter	30.7	34.83

2.5 IGNITION OF SAMPLES

Several techniques (paper, cardboard, and electric coil) were tried in an attempt to ignite the samples. By far the best method to use was a Meker Blast Burner equipped with an air line. Ignition of all samples with this system occurred within several minutes.

2.6 CHAMBER SAMPLING

Teflon sample lines (0.6 cm:OD) for each instrument were placed within the center of the chamber at a height 1.5 m above the floor.

2.7 QUALITY ASSURANVE OF EXPERIMENTAL MEASUREMENTS

The gas monitors were typically calibrated after every fourth analysis throughout the study with commercially available standards. The following gases were used:

CO: 47.48 ppm Scott Specialty Gases

SO_2: 0.63 ppm Scott Specialty Gases

NO_2: 0.86 ppm Matheson Gases

POC: Hydrocarbon Free Air Scott Specialty Gases (for instrument zeros)

POC: 0.951 ppm Toluene in Hydrocarbon Free Air Scott Specialty Gases

2.8 PARTICULATE SAMPLER

The Harvard respirable particulate sampler used in this study is an active integrating sampler with a cut-size of 2.5 μm. Only particulates smaller than 2.5 μm in diameter are collected on a filter, which is subsequently weighed.

The sample flow of the Harvard Respirable Particulate Sampler was calibrated before and after each analysis using a calibrated flow tube. The respirable particulate filters were weighed on a Metler AE 163 analytical balance.

3. EXPERIMENTAL PROCEDURE

3.1 SAMPLE PREPARATION

A random sample of each type of charcoal was broken into small chunks (1 to 2 cm in height and thickness). Fifty grams of the broken sample were then placed in a small pyramid (no more than three chunks high) in the center of the Chimney Quick apparatus.

3.2 SAMPLE IGNITION

The Chimney Quick apparatus containing the sample was held over a hot gas/air flame for several minutes. Ignition was confirmed by inspection of the samples from the bottom of the apparatus. After ignition was confirmed, the apparatus was placed in the chamber for emission measurements.

3.3 MONITORING PERIOD

Combustion was generally complete after about one hour; however, the measurements were continued an additional three to four hours to measure the removal of pollutants from the chamber (~3 chamber volumes) and to determine the air exchange rate of the chamber. After the chamber had returned to preignition conditions, the ashes were collected and weighed.

3.4 PARTICULATE SAMPLING

The Harvard respirable particulate sampler and a pump capable of drawing 10 L/min of air through the sampler were placed inside the chamber

near the center at a height of 1 m from the floor. After the fuel sample was placed in the chamber, the Harvard respirable particulate sampling pump was turned on and the flow measured with a calibrated flow tube. The particulate sampler was then allowed to sample for 4 h. After sampling was completed, the flow of the particulate pump was remeasured, the pump was turned off, and the filter was removed and weighed.

3.5 AIR EXCHANGE MEASUREMENTS

The air exchange rate of the chamber was measured several times during the investigation by monitoring of CO decay with respect to time. This was performed two ways: (1) CO was manually injected into the chamber and decay rate measured, and (2) the decay of CO was monitored after sample combustion was completed. Based on these measurements, the average air exchange rate for the period of investigation was determined to be $0.78 \pm 0.034\ h^{-1}$.

3.6 DATA ANALYSIS AND CALCULATIONS

After a minimum of four good combustion experiments per sample (except for the PMDC briquettes), the data were collected and analyzed by computer (IBM PC) using a spreadsheet program (Lotus 1-2-3) for each of the measured experimental parameters. Plots were generated (mg/m^3) vs elapsed time to inspect the data for experimental problems. Due to instrumental problems some of the experimental runs do not have an entire set of measurements for each day. A numerical average of the good experimental data is reported. Figure 1 shows the CO concentration in the chamber for replicate experiments. Based on these profiles, we believe that a single average value adequately represents the data. When measurements are made for less than 6 h, the "tail" of the distribution, which is due to exhausting of the pollutants from the chamber following the completion of combustion, is extrapolated from the measured data based on the exponential decay in pollutant concentration. At least 2 h of measurements are made prior to extrapolation.

The following calculations were made in analyzing the averaged data:

Emission Factor (EF): $V \cdot ACH \cdot dT \cdot \int C/W$

Respirable Suspended Particulate (RSP): RSP: $EF = [(W_f - W_i) \cdot V]/(W \cdot F \cdot T)$

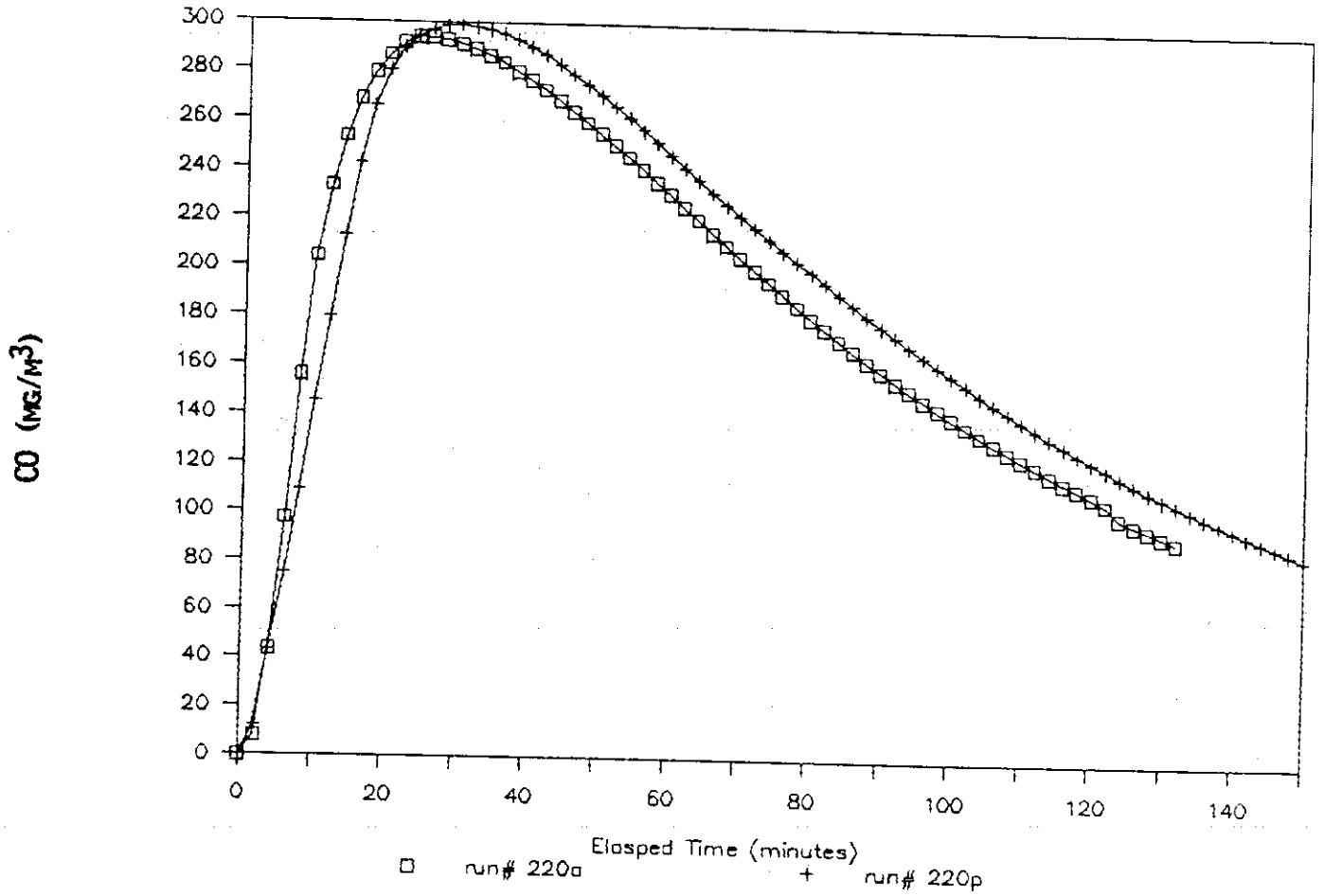

FIGURE 1. COMPARISON OF CO CONCENTRATION FROM REPLICATE EXPERIMENTS.

Combustion Efficiency (CF): $CF(\%) = (1 - A/W)(100\%)$

Where

V = Chamber volume (m^3) - 17 m^3

ACH = Air exchange rate - 0.78 h^{-1}

dT = Sampling interval (h)

$\int C$ = Integrated concentration of a specific pollutant (g/m^3)

A = Ash weight (g)

W_i = Initial filter weight (g)

W_f = Final filter weight (g)

W = Fuel weight (kg) - typically 0.05 kg

F = Flow of RSP sampler (m^3/min)

T = Sampling time of RSP sampler (min)

4. RESULTS AND DISCUSSION

4.1 COMBUSTION EFFICIENCY

The average combustion efficiencies for the different fuels are listed in Table III. As can be concluded from the data, natural wood charcoal has the highest combustion efficiency of the fuels tested. The other samples contain additives that do not burn; for example, the PMDC briquettes had a large amount of cinder content after combustion. Placing this material directly on the burner for 1-h did not produce a significant decrease in mass. We therefore conclude that the PMDC briquettes contain an inert substance that does not combust.

Table III. Combustion efficiency of the samples tested

Sample type	Combustion efficiency (%)		Ash description
	Average	Range	
Pakistani	95.4 ± 1.3	94.2-96.7	Light, fluffy
Barbecue	77.4 ± 2.4	73.8-79.2	Grainy
Charania	76.2 ± 0.9	75.2-77.8	Grainy
PMDC*	24		Cinders

*Only one analysis performed.

Another problem associated with the combustion of PMDC briquettes was the inability to sustain fuel sample combustion. A total of four attempts were made to ignite and sustain combustion of the fuel sample. The first three attempts extinguished within 1 to 2 min after removal from the burner. The fourth attempt produced a marginally sustaining combustion of approximately 9 min duration. The combustion, although short and incomplete, was sufficient to yield some useful data. These results are discussed below.

4.2 SUSPENDED PARTICULATES

The type of particulate sampler used in this study, the Harvard Respirable Particulate Sampler, measures only particulates smaller than 2.5 μm. Suspended particulate emission factor data for the samples tested are presented in Table IV.

Table IV. Emission factors for respirable particulates (<2.5 μm) for different fuels

Sample type	Average (g/kg)	Range (g/kg)	Appearance of filter
Pakistani	0.243 ± 0.078	0.188-0.299	Dark Tan
Barbecue	0.153 ± 0.067	0.086-0.220	Light Tan
Charania	0.141 ± 0.042	0.088-0.184	Dark Tan
PMDC*	0.384		Black

*Based on only one analysis.

From the results shown in Table IV, several general conclusions can be drawn: (1) combustion of Charania briquettes, barbecue briquettes, and Pakistani wood charcoal yields similar amounts of respirable suspended particulates, and (2) the PMDC briquettes generate substantially higher levels of RSP (however, only one sample of PMDC was run).

4.3 EMISSION OF CARBON MONOXIDE, SULFUR DIOXIDE, NITROGEN OXIDES, AND PHOTOIONIZABLE ORGANIC COMPOUNDS

The two variables of interest in this study are: (1) peak concentration and (2) emission factor. The peak concentration can vary due to such experimental factors as initial sample weight, the air exchange rate of the chamber, and the combustion rate of the sample. Clearly, the first two of these experimental factors can be controlled or held constant. However, the combustion rate of the sample is more difficult to control since it can be a function of many different variables such as the sample and the sample pyramid shape. In addition, the rate of combustion can be a function of the extent of initial sample ignition. Therefore, the peak concentration can be used only as a reference for relative emissions.

The emission factor, however, normalizes the experimental variables of air exchange, sample weight, and combustion rate because it is a measurement of the total mass of emissions generated per unit mass of dry fuel consumed. Therefore, for comparison purposes between different fuels, the emission factor is the best choice.

In order to measure meaningful concentrations and emission factors for the gases of interest, five key assumptions were made: (1) sufficient oxygen was present within the chamber to assure a steady and complete burn; (2) the relative humidity and temperature had no effect on the combustion process and the monitoring of the gases of interest; (3) air flow within the chamber was sufficient for good mixing; (4) no sink effects were present within the chamber to decrease the monitored concentration; and (5) the amount of SO_2 and NO_x lost to the moisture on the chamber walls was constant.

Of these assumptions only the last two, 4 and 5, could not be measured or experimentally observed during this study. In order to validate assumptions 1 and 2, the following parameters were kept constant throughout the study: (1) fresh air was injected into the chamber at a constant rate of 180 L/min, and (2) the temperature and relative humidity were kept constant at 15.0 °C and 20%, respectively. The assumption of good intra-chamber mixing was validated with the following experiment.

With the mixing fans on, CO was injected into the chamber and sampling was performed from various locations to measure instrument response. The results indicated less than a 5% variation between sample location and sample height within the chamber. Sink effects are discussed subsequently; however, these may be significant for some of the pollutants. Consequently, actual emission factors may be somewhat higher than reported emission factors due to these losses.

4.3.1 CO Emissions

Figure 2 and Table V illustrate the average time-dependent CO concentration and the emission factor observed for the various fuels, respectively. The Pakistani charcoal, the barbecue briquettes and the Charania briquettes peaked at very similar concentrations. However, upon a closer examination, the emission factor of the Charania briquettes was 32% higher than the emission factor of the Pakistani charcoal and is very close to the barbecue briquettes in magnitude. This is attributed to the fact that the Charania briquettes, like the barbecue briquettes, were experimentally observed to be a much slower burning substance than the Pakistani charcoal. The observation that the Pakistani charcoal burns more rapidly than the Charania briquettes is confirmed in Figure 2.

Table V. Peak CO concentrations and emission factors for the tested fuels

Sample type	Peak CO (mg/m^3)	Time maximum peak (h)	Emission factor (g/kg)
Pakistani	325	0.467	169
Barbecue	387	0.867	251
Charania	335	0.800	223
PMDC*	108	0.367	159

*Based on one analysis.

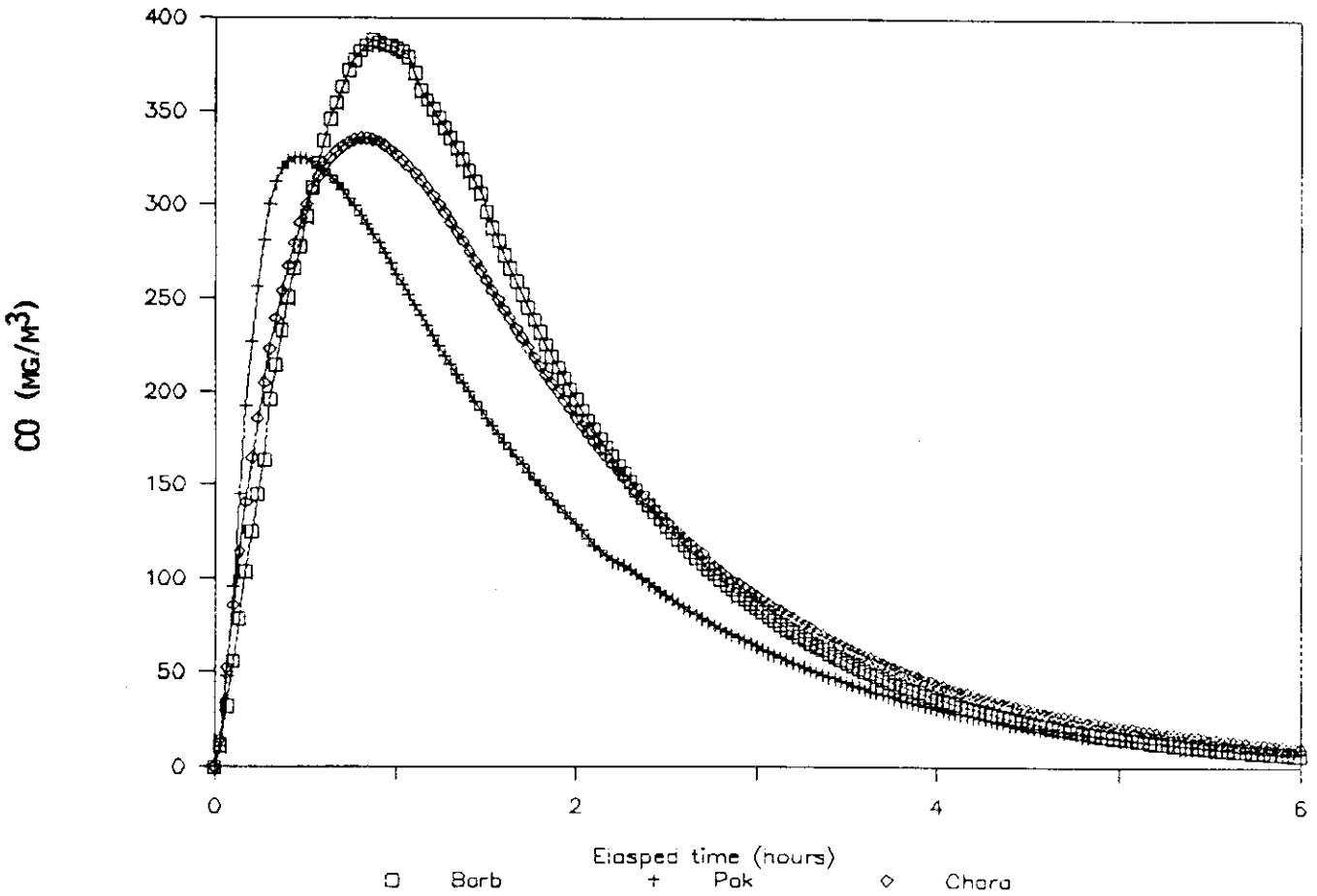

FIGURE 2. AVERAGE CO CONCENTRATIONS FOR TESTED FUELS.

4.3.2 Photoionizable Organic Compound Emissions

The instrument being used in this study for POC measurements, the HNU, works on the principle of photoionization. Chemical species with an ionization potential equal to or less than the output of the photoionization tube will undergo ionization and be detected. Species with a greater ionization potential will not be detected. For this study, the ionization potential of 10.2 eV was selected. At this ionization potential, NO and NO_2 will undergo photoionization. However, their sensitivity to photoionization is approximately a factor of 100 less than that of toluene. Therefore, the contribution of NO and NO_2 to the POC concentration is expected to be small.

Different organic species have different photoionization potentials and sensitivity. Because it would be impossible to calibrate the HNU with all possible combustion products, we have chosen toluene (I.P. 8.82 eV) as the standard response for all POC measurements. The POC results reported in Table VI, therefore, are expressed in toluene equivalent units.

Table VI. Photoionizable organic compounds peak concentrations and emission factors for tested fuels*

Sample type	Peak POC (mg/m^3)	Time maximum concentration (h)	Emission factor (g/kg)
Pakistani	10.3	0.333	4.38
Barbecue	10.8	0.267	4.73
Charania	12.5	0.267	5.73
PMDC†	6.4	0.367	7.43

*Based on Toluene Equivalent Units.
†Based on one analysis.

The results in Table VI and Figure 3 show that the organic pollutants reach maximum concentration faster than CO. A comparison of the time to reach maximum concentration for CO (Table V) and POC (Table VI) reveals that, overall, maximum organic concentrations occur in approximately half the time of maximum CO concentrations. In addition, unlike CO, all fuel

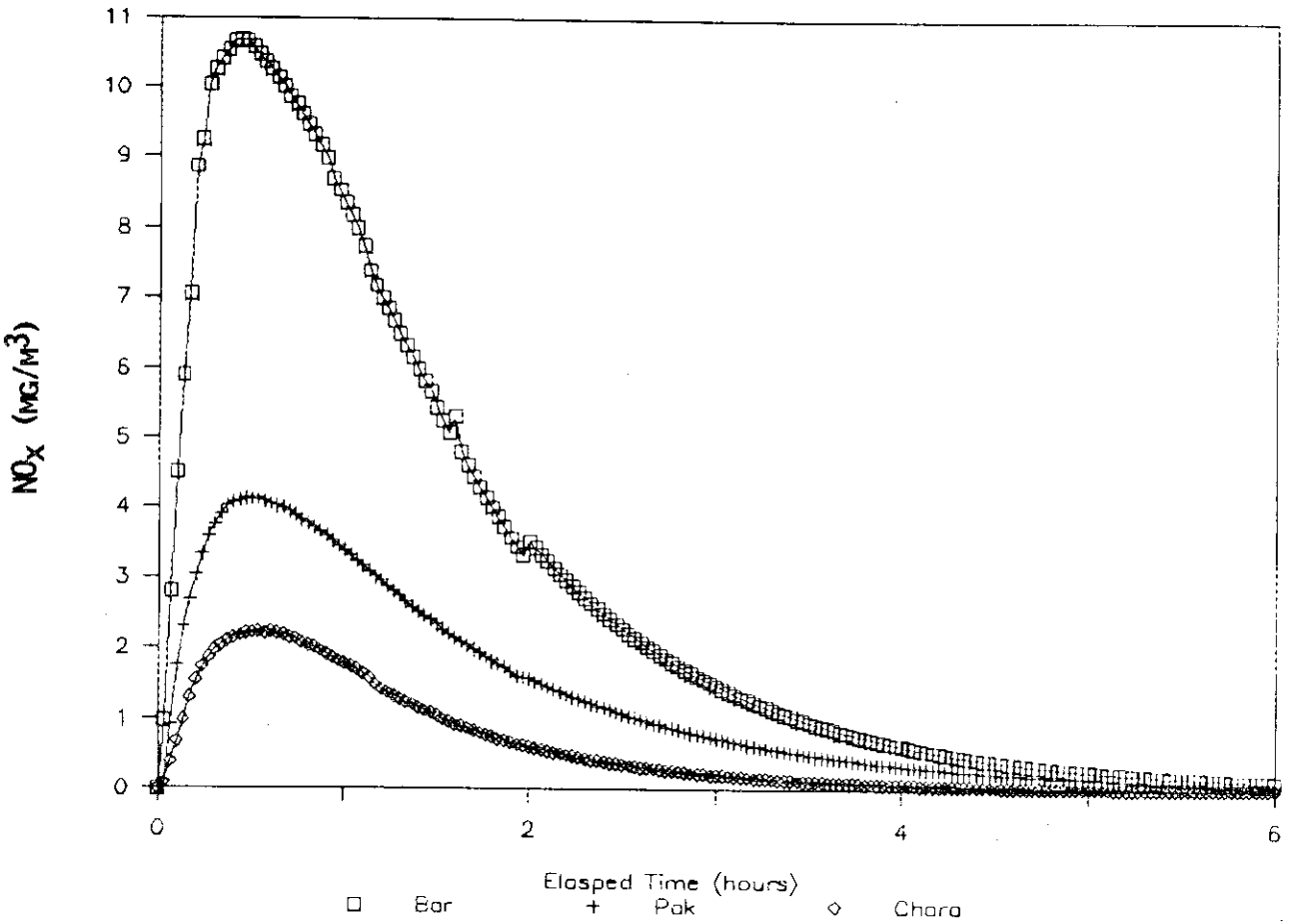

FIGURE 3. AVERAGE PHOTOIONIZABLE ORGANIC COMPOUND RESPONSE FOR TESTED FUELS.

types reached the maximum POC concentrations at about the same time (Figure 3). This might be expected because, as the sample heats up, organics will volatilize and be emitted by the sample. CO, on the other hand, is a product of combustion and its emission is determined solely by the rate of combustion. More study of organic emissions is required before any definite conclusions can be drawn about their potential for producing adverse health effects.

The Charania fuel is about 20% higher in POC than the Pakistani charcoal and the barbecue briquettes. This increase in POC may be due to the presence of molasses in the Charania briquettes. An interesting experiment would be to combust a sample of Charania briquettes without the molasses to see if the POC decreased to the same concentrations as the other samples.

4.3.3 NO_x Emission

The nitrogen oxides measurements are slightly more complex than the CO and POC measurements. The first problem is associated with the unknown chamber sink effects and the loss to wall moisture for these reactive compounds. The second problem is more complex due to the two possible types of combustion products, NO and NO_2. Because of instrument and calibration problems, we were unable to measure the gases separately, only together. Therefore, for calculation purposes, we have assumed that the bulk of the NO_x generated was NO. We feel that this assumption is valid since, in an oxygen-deficient environment, nitrogen oxide would be the dominant species. Therefore, the results reported in Figure 4 and Table VII are reported in NO equivalent units.

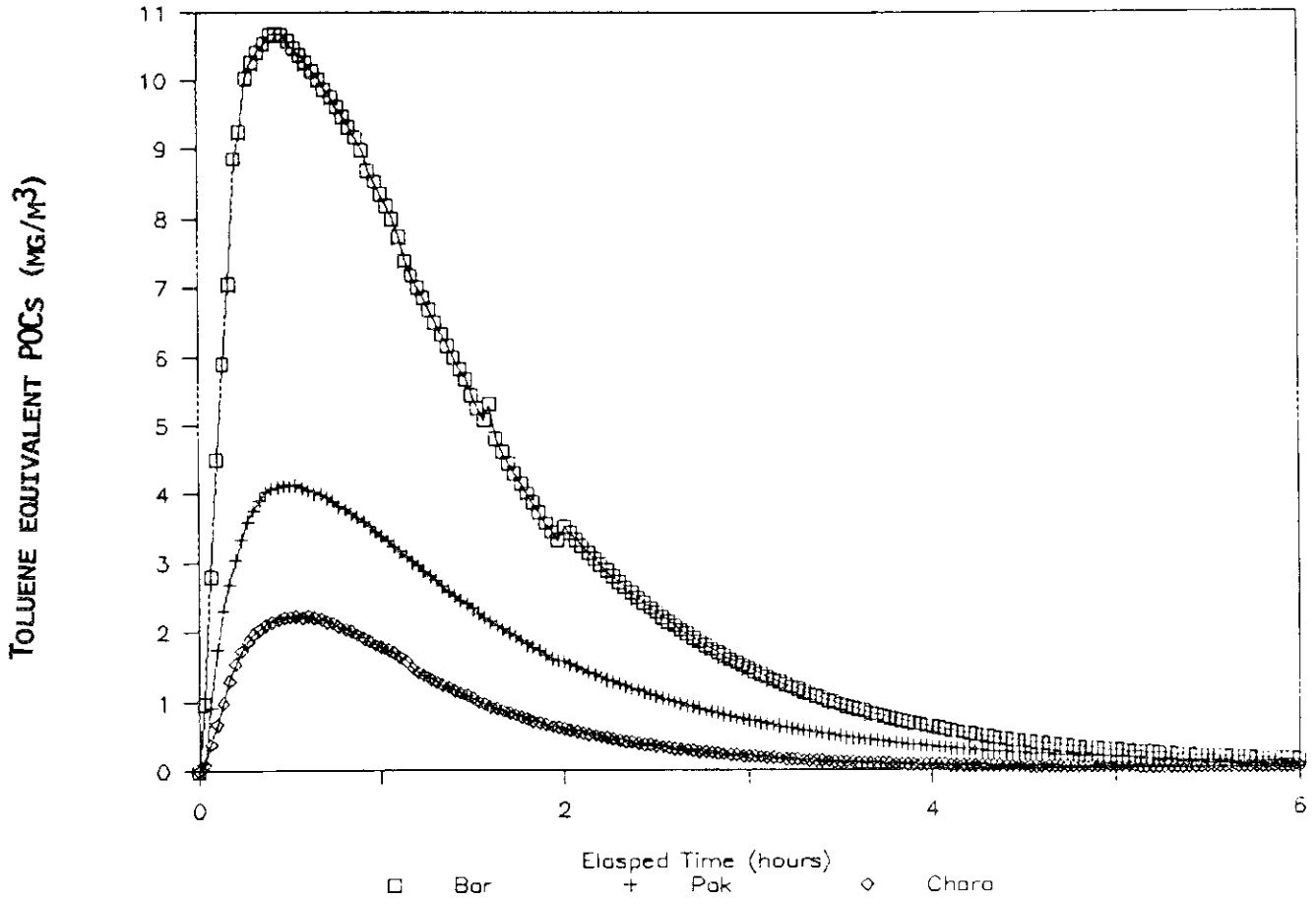

FIGURE 4. AVERAGE NO_X CONCENTRATIONS FOR TESTED FUELS.

Table VII. Nitrogen oxide concentrations and emissions factors for the test samples*

Sample type	Peak NO concentrations (mg/m^3)	Time maximum concentrations (h)	Emission factor (g/kg)
Pakistani	4.1	0.467	2.09
Barbecue	10.7	0.433	5.87
Charnia	2.2	0.600	1.10
PMDC†	1.7	0.333	1.71

*Based on Nitrogen Oxide Equivalent Units.
†Based on one analysis.

As can be concluded from the data in Table VII, the highest concentration of NO is found with commercial barbecue briquettes. The fact that these briquettes are a factor of 3 to 6 times higher in concentration is not surprising since sodium nitrate is a common additive within barbecue briquettes. The NO emission factor for Charania briquettes is approximately half that of the Pakistani wood charcoal. This effect may be due to the faster (and presumably hotter) combustion of the Pakistani charcoal, although further investigation of wall loss effects are in order before final conclusions are made.

4.3.4 SO_2 Emission

Like the NO_x measurements, the SO_2 measurements are susceptible to unknown sink effects and losses of SO_2 to moisture in the chamber. In fact, the relatively low SO_2 concentration makes wall losses of greater concern than for NO_x. In addition, the SO_2 concentration was generally only slightly above the instrument noise level (with the exception of the PMDC briquettes experiment, where the concentration was quickly off scale of the instrument used, >5 ppm). Therefore, the results are only semiquantitative. Table VIII gives the SO_2 results obtained.

Table VIII. Sulfur dioxide peak concentrations and emission factors for the tested samples

Sample type	Peak SO_2 (mg/m^3)	Time maximum conc. (h)	Emission factor (g/kg)
Pakistani	0.280	0.133	0.127
Barbecue	0.410	0.233	0.125
Charania	0.267	0.267	0.229
PMDC	>12.5		

As can be concluded from the data in Table VIII, the emission factor for SO_2 of the Charania briquettes is approximately a factor of 2 higher than the Pakistani wood charcoal. This higher concentration of SO_2 is consistent with the fact that the Charania briquettes consist of 20% coal. Additional measurements will be required to determine the emission factor with greater confidence. However, based on the initial experiment, the PMDC briquettes will have a substantially higher emission factor for SO_2 than the other fuels.

5. CONCLUSIONS

The results from this limited study suggest that the Charania briquettes are somewhat higher in CO, SO_2, and POC than the Pakistani wood charcoal, but are similar in RSP and lower in NO_x concentrations. However, to a first approximation, the pollutant emissions of the Charania briquettes are similar to the currently used material (Pakistani wood charcoal). Additional study is needed to obtain more refined estimates of combustion emissions of the various fuels.

In addition, we have demonstrated that the environmental chamber can be used to simulate the burning of fuel samples with a nonvented stove within a closed environment. The chamber offers the capability to study the emissions of fuel samples at various air exchange rates, temperatures, and humidities.

These results, consistent with other limited measurements in developing countries, do, however, indicate that unvented combustion for cooking and heating can produce levels of pollutants that may be of concern from an acute health perspective and that are quite likely to produce chronic health effects.

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