

Research and Development

GREENHOUSE GASES FROM SMALL-SCALE
COMBUSTION DEVICES IN DEVELOPING
COUNTRIES: CHARCOAL-MAKING
KILNS IN THAILAND

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FOREWORD

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GREENHOUSE GASES FROM SMALL-SCALE COMBUSTION DEVICES IN DEVELOPING COUNTRIES

Charcoal-Making Kilns in Thailand

by

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FOREWORD

This work is part of a multi-year international study of the greenhouse-gas implications of small-scale combustion devices in developing countries. To date, measurements have been made of household stoves in India and China and charcoal kilns in Thailand. Although individually small, these devices are so numerous and their emission factors per unit output are so significant that, in total, they can have an appreciable influence on global and national inventories of important greenhouse gases.

The charcoal kiln measurements described in this report took place at the Charcoal Research Centre (CRC) in Saraburi, Thailand, which is operated by the Royal Forest Department. In Thailand, overall management was done by Khummongkol; Pennise, Chaiwong, and Ritgeen took the samples and conducted lab analyses; and Panyathanya and his CRC team operated the kilns. In the USA, Rasmussen, Khalil, and Zhang were responsible for lab analyses and quality control. Smith was overall manager of the project. Overall financial and administrative management of the project took place at the East-West Center, funded though a Cooperative Agreement with the USEPA, overseen by Thorneloe.

ABSTRACT

Airborne emissions from charcoal-making kilns commonly used in the developing world were measured during typical operating conditions. The kilns tested were of five types: brick beehive, mud beehive, earth mound, rice husk mound, and single (oil) drum. These experiments were carried out in Thailand, a nation which produces about 7.2 million tons of charcoal per year of the estimated 26-100 million tons produced globally. Emission factors for the production of charcoal were determined for the direct greenhouse gases carbon dioxide ($\rm CO_2$), methane ($\rm CH_4$), and nitrous oxide ($\rm N_2O$), the indirect greenhouse gases carbon monoxide ($\rm CO_2$) and total nonmethane hydrocarbons ($\rm TNMHC$), as well as total suspended particulates ($\rm TSP$). Charcoal production efficiency (yield) and charcoal and fuelwood composition were determined as well.

As is generally known to be the case for charcoal making, the conversion of wood carbon to charcoal carbon was fairly inefficient, ranging from a low of 48% for the earth mound kilns to a maximum of 57% for the more efficient brick beehive kilns. Average emission factors, expressed as grams of pollutant per kilogram of charcoal produced, for the three runs of each of the five kiln types ranged from 970-1600 for CO_2 , 13-58 for CH_4 , 110-340 for CO_2 , 9-95 for TNMHC, 0.017-0.084 for N_2O_2 , and from 0.7-4.2 for TSP. Hence, a substantial fraction of the original fuel carbon was lost as CO_2 and other products of incomplete combustion (PIC). On average, fuelwood carbon is approximately diverted as follows: 52% to charcoal, 24% to CO_2 , and 10% to PIC. Thus, due to the higher global warming potentials of PIC relative to CO_2 on carbon basis, such kilns can produce rather large net greenhouse gas emissions, even when the wood is harvested renewably.

CONTENTS

	<u>Page</u>
Foreword	ii
Abstract	ii
List of Figures	iv
List of Tables	V
Glossary	vi
Acknowledgments	vi
I. Introduction and Summary	1
II. Conclusions and Recommendations	3
III. Methods	
A. Kilns and Wood Tested	4
B. Parameters Measured	4
C. Summary of Experimental Procedure	5
IV. Results	
A. Airborne Sampling Measurements	9
B. Solid Product Measurements	9
C. Carbon Balance and Calculation of Emission Factors	16
D. GC Analysis of Canister Samples	20
E. Grab Samples for Determining Emission Ratios and Emission	
Factors	20
V. Discussion	28
VI. References	35
Appendix A: Construction and Operation of the Charcoal Kilns	37
Appendix B: Detailed Sampling Procedure	47
Appendix C: Analytical Methods	51
Appendix D: Follow-up Experiments to Determine Condensable Liquid	
Emissions	52
Appendix E: Quality Assurance Plan	57

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
Figure 1.	A schematic of the hood and duct system used for emissions sampling in the 15 kiln experiments	7
Figure 2.	Distribution of the original wood carbon in the products of the charcoal-making process	19
Figure 3a-c.	Comparison of net emission concentration ratios for KMUTT data and OGIST data	23
Figure 4.	Carbon cycle for charcoal making in the BBH kiln and the GWC resulting from renewable and non-renewable harvesting of the wood	31
Figure 5.	Carbon cycle for charcoal making in the SD kiln and the GWC resulting from renewable and non-renewable harvesting of the wood	32
Figure A-1.	Diagram of the brick beehive kiln (reproduced from Chomcharn, 1985)	40
Figure A-2.	Brick beehive kiln at the Charcoal Research Centre	40
Figure A-3.	Diagram of the mud beehive kiln (reproduced from	
C	Chomcharn, 1985)	41
Figure A-4.	Ignition of the mud beehive kiln in the hood system at the Charcoal Research Centre	41
Figure A-5.	Mud beehive kiln inside the hood system at the Charcoal	
8	Research Centre	42
Figure A-6.	Diagram of the single drum kiln (reproduced from	
8	Chomcharn, 1985)	43
Figure A-7.	Single drum kiln at the Charcoal Research Centre	44
Figure A-8.	Diagram of the earth mound and rice husk mound kilns	
S	(reproduced from Chomcharn, 1985)	45
Figure A-9.	Ignition of the rice husk mound kiln at the Charcoal Research	
S	Centre (reproduced from Chomcharn, 1985)	46
Figure B-1.	A schematic of the sampling system used in the 15 kiln	
J	experiments	48
Figure D-1.	A schematic of the sampling system used in the follow-up	
_	experiments	56

LIST OF TABLES

No.	<u>Title</u>	<u>Page</u>
Table 1.	Sampling summary	6
Table 2.	Emission sample concentrations (from KMUTT mixing bag	
	data) and ambient concentrations	10
Table 3.	Net emission ratios of gases and TSP to CO ₂	11
Table 4.	Solid product measurements	12
Table 5.	Carbon analyses and calorific values of solid products	14
Table 6.	Charcoal and brands yields (mass, carbon, and energy bases)	15
Table 7a-d.	Average emission factors (a. g pollutant/kg charcoal produced;	
	b. g pollutant/kg dry wood; c. g pollutant carbon/kg charcoal	
	carbon produced; d. g pollutant carbon/kg wood carbon)	17
Table 8.	Percent distribution of the original wood carbon in the	
	products of the charcoal making process	18
Table 9.	Comparison of emission ratios of CO, CH ₄ , and TNMHC to	
	CO ₂ determined using OGIST canister data and KMUTT	
	mixing bag data	22
Table 10.	Comparison of emission factors determined using OGIST	
	canister data and KMUTT mixing bag data (g pollutant/kg dry	
	wood)	24
Table 11.	Comparison of grab sample method to mixing bag method:	
	emission ratios of CO, CH ₄ , and TNMHC to CO ₂	25
Table 12.	Comparison of emission factors determined using grab sample	
	method and mixing bag method (g pollutant/kg dry wood)	26
Table 13.	Summary of previous charcoal-making kiln emission studies	30
Table 14a-b.	Estimated annual air pollution emissions from charcoal	
	production in Thailand, 1996 (Total and carbon only)	33
Table 15.	Estimated global warming commitments (GWC) from Thai	
	and world charcoal production compared to GWC from fossil	
	fuel cycle	34
Table D-1	Data from follow-up experiments (determination of	
	condensable liquid emissions)	54
Table E-1	Replicates: Two bags filled simultaneously through a y-probe	59
Table E-2	Storage test data	60
Table E-3	Analyses of duplicate small bags filled from the same large	
	mixed bag	61
Table E-4	Field and laboratory blank filter data	61

GLOSSARY

BBH brick beehive kiln

CRC Charcoal Research Center

EM earth mound kiln

EOHSI Environmental and Occupational Health Sciences Institute

FID flame ionization detector GC gas chromatography GHG greenhouse gas

GWC global warming commitment GWP global warming potential

HC hydrocarbons

KMUTT King Mongkut's University of Technology Thonburi

MBH mud beehive kiln Mt megatons (metric)

MtC megatons of carbon (metric)

OGIST Oregon Graduate Institute of Science and Technology

PIC products of incomplete combustion ppb parts per billion (by volume or mole) ppm parts per million (by volume or mole)

RHM rice husk mound kiln SD single drum kiln SM sawdust mound kiln THC total hydrocarbons

TNMHC total non-methane hydrocarbons
TNMOC total non-methane organic compounds

TSP total suspended particulates VOC volatile organic compounds

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I: INTRODUCTION AND SUMMARY

Biomass burning plays important roles in the global carbon cycle. Although complete combustion of biomass produces little more than CO₂ and water, most actual combustion is done in circumstances that result in substantial diversion of biomass carbon into products of incomplete combustion (PIC). Indeed, current estimates are that biomass combustion accounts not only for 25-45% of the annual global emissions of CO₂, but also for 15-50% of CO, 3-10% of CH₄, and 24% of total non-methane organic compounds (TNMOC) (Levine, 1990, Crutzen and Andreae, 1990, Andreae, 1991). CO₂ and CH₄ are in addition the two most important greenhouse gases (GHG) and CO and TNMOC indirectly affect global warming through atmospheric chemical reactions that in turn affect GHG levels. A good characterization of biomass burning thus is important for achieving scientific understanding of the potential for human activities to engender global warming, as well as informing the international political/economic discourses about what GHG mitigation measures are warranted and who should pay for them.

Combustion of biomass harvested or naturally regrown on a sustainable basis does not cause a net increase of CO₂ in the atmosphere. Unfortunately, through deforestation and other non-renewable practices, much burned biomass is not replaced. Even with complete recycling of the carbon, however, a biomass fuel cycle can produce a net increase in global warming commitment (GWC) because of the emitted PIC, which have, on average, a higher global warming potential (GWP) per kilogram carbon than CO₂. As a result of these two factors, partially non-renewable harvesting and significant PIC production, there has been much work in recent years to characterize biomass combustion of different kinds in different seasons around the world (Levine, 1996).

Most biomass combustion, whether natural or anthropogenic, is done in circumstances in which access to air is not greatly restricted. Indeed, for most human uses, better combustion efficiency is a distinct advantage. Thus, although not perfect, combustion efficiency is normally relatively high, i.e., only a few percent of fuel carbon is diverted to PIC. There is at least one major exception to this pattern, however, charcoal production, which is done basically by heating the fuelwood in the absence of air, thus creating a higher quality fuel, but at the expense not only of a significant loss of energy in the starting fuelwood, but also significant production of PIC.

The Food and Agriculture Organization reports that about 26 million tons (Mt) of charcoal were produced worldwide in 1995 (FAO, 1997) occurring largely in the developing world. This represents about 12% of worldwide fuelwood use (FAO, 1997). Other estimates of the amount of charcoal produced annually worldwide include 100 million tons (Rosillo-Calle et al., 1996). The lower end estimates may not include all of the production of charcoal in the noncommercial sector, explaining some of the rather large range in the estimates. Charcoal production has increased at a rate of approximately 3% per year over the period 1991-1995 (FAO, 1997). Because of its large PIC production, it might be expected that charcoal's impact on GWC is substantially greater than its share of fuel demand. To date, however, the airborne emissions from charcoal-making are poorly characterized in existing greenhouse gas emission databases.

The charcoal kilns used in the developing world are not easily monitored, because they often operate in remote areas over many days or even weeks for a single run. Emissions vary dramatically over the run and may be released from a number of locations on the kiln, which can be nothing more than a carefully constructed mound of earth many meters long.

This project was designed to characterize emissions from the most common charcoal kilns in the developing world. In this phase, we worked at the Charcoal Research Centre (CRC) in Thailand, the Asian country with the largest charcoal production. Although challenging enough, conditions at the CRC were more controllable than would be the case at a commercial kiln operating in the forest. Thus, it provided a good location for developing and validating methods to be used later in Africa and Latin America.

In addition, Thailand's charcoal production itself is substantial. Some 7.2 million tons of charcoal are produced per year (Thailand Department of Energy Development and Promotion, 1996). Charcoal accounts for 9% of Thailand's total energy consumption, and 98% of use and production takes place in rural areas (Thailand Department of Energy Development and Promotion, 1996). Thus, Thai kilns could potentially account for a significant part of overall Thai GHG emissions.

We found that average emission factors, as grams of pollutant per kilogram of charcoal produced, over three runs of each of the five kiln types, ranged from 970-1600 for CO_2 , 13-58 for CH_4 , 110-340 for CO, 9-95 for TNMHC, 0.017-0.084 for N_2O , and from 0.7-4.2 for TSP. This means fuelwood carbon is approximately diverted as follows: 52% to charcoal, 24% to CO_2 , and 10% to PIC. Put another way, even if the charcoal were burned with absolutely no PIC production during its final enduse, nearly one-fifth of the carbon available in the fuelwood has already been released as PIC during charcoal production.

Based on published GWPs (using a 20-year time horizon) for CH₄ and N₂O only, we estimate that 0.65-1.41 kg C-CO₂ (carbon as carbon dioxide equivalents) is emitted per kg charcoal produced. Based on reported distributions of production among kiln types, we estimate that the total primary GWC of Thai kiln emissions is about 5.0 Mt C-CO₂. This is approximately 7.4% of total Thai emissions from fossil fuel combustion. If our results accurately reflect kiln performance worldwide, some 20 Mt C-CO₂ comes from kilns (assuming an annual global production of 26 Mt of charcoal), which equals about 0.3% of that from global fossil fuel consumption (IPCC, 1995). (If GWPs above 1.0 are adopted for the other PIC emitted during charcoal making, these numbers would be higher.) Total emissions from the entire charcoal fuel cycle, of course, would also include the emissions from the stoves and other devices in which the produced charcoal is burned.

At the conclusion of this report, we compare our measured emission factors to the results of previous studies and describe how our methods might be applied to kilns in less-controlled circumstances.

II: CONCLUSIONS AND RECOMMENDATIONS

Simple charcoal kilns of the types commonly used in developing countries convert only some 43-47% of the energy content of wood into charcoal. Our measurements in Thailand indicate that, in the process, Thai kilns lose 40-50% of the wood carbon, approximately distributed as follows: 20-25% as CO₂; 9-18% as gaseous products of incomplete combustion (PIC-CO, CH₄, HC); and the remainder as ash, aerosol, and brands (partially carbonized solids). As a result of this significant diversion of carbon to PIC, which have higher global warming potentials (GWP) than CO₂, the overall global warming commitment (GWC) of the charcoal fuel cycle is significantly higher than that from the CO₂ alone. For example, if the wood is harvested non-renewably (the carbon is not recycled), the GWC of the kiln is 1.9-3.6 times the GWC of the charcoal burned at the stove, assuming that the stove charcoal is completely converted to CO₂. The GWC of non-renewable charcoal fuel cycle, therefore, is some 3.9-6.2 times that of a fossil fuel cycle producing the same energy (assuming that the fossil-fuel GWC previous to final combustion is 10% of the total for the fossil-fuel cycle). Even if the wood is harvested renewably (complete carbon recycling), the GWC of the charcoal fuel cycle is still some 2-4 times greater than produced by burning an equivalent energy content of liquid or gaseous fossil fuel. (These calculations account for the GWP of CH₄, CO₂, CO and HC. If GWPs of only the first two are counted, the values decrease by about 50%.)

This implies that charcoal fuel cycles are among the most greenhouse-gas (GHG)-intensive in the world. A natural gas fuel cycle, for example, would have to leak directly into the atmosphere some 12-25% of the CH₄ it delivered for combustion to emit as much GHG per unit delivered energy as a non-renewable charcoal fuel cycle.

These emission factors can be applied to other areas of the world where similar charcoal-making methods are used. This will allow for somewhat better global estimates of the inventory of greenhouse gas and air pollutant emissions from the production of charcoal. More localized emissions sampling is necessary, however, for accurate determination of emission factors for two reasons. First, even given the same kiln type, there is great variability globally in both kiln sizes and construction techniques. Second, kiln behavior is largely dependent on operator tending methods, which again vary greatly around the world and can even vary within the same operator across different charcoal making sessions.

The results of this study are consistent with the default values listed by IPCC for charcoal kilns in that the IPCC values fall within the range of the kilns monitored here. The IPCC values are perhaps closest to those of the brick beehive kiln in Thailand. Depending on the kiln type, however, compared to the IPCC values our values range from -2x to +1.6x for CO; -1.4x to +1.9x for CH₄; and -6.0x to +1.9x for TNMOC. Thus, to accurately reflect actual emissions, there is need to specify the fraction of charcoal produced in different major kiln types.

III: METHODS

A. Kilns and Wood Tested

Based on knowledge of the charcoal kilns commonly used in Thailand, five types were selected for testing (Chomcharn, 1985):

- <u>Brick Beehive</u> (BBH): beehive-shaped and charged with about 800 kg wood with a firing time of about 40 hours. It is a popular, improved version of the MBH kiln.
- <u>Mud Beehive</u> (MBH): somewhat smaller using about 600 kg wood per run with a firing time of about 40 hours.
- <u>Earth Mound</u> (EM): layers of grass, leaves, and a final layer of dirt over a charge of about 200 kg wood taking about 20 hours. This type is common throughout the developing world.
- Rice Husk Mound (RHM): Similar to EM but with layer of rice husks plus 200 kg wood but taking only about 3 hours. Most commonly used in agricultural areas.
- <u>Single Drum</u> (SD): made from an oil drum and using about 80 kg wood with a firing time of about 4 hours. Portable and cheap.

On-site staff of the Thai Royal Forest Department built, operated, and maintained the kilns at the Charcoal Research Centre (CRC) in Saraburi, Thailand. All of the kilns were built under a large shed with a metal roof and open sides to allow research in all weather conditions.

Two species of wood commonly used for charcoal production, *Eucalyptus camaldulensis* and *Leucaena leucocephala*, were chosen for these charcoal kiln tests. Three runs were conducted for each kiln type, where runs 1 and 2 used eucalyptus and run 3 used leucaena. Drawings, photos, and other details of the construction and operation of the kilns are in Appendix A.

B. Parameters Measured

Since the purpose of the sampling was to determine a detailed carbon balance for the kilns as well as to quantify greenhouse-gas emissions, nearly all inputs and outputs were monitored.

- <u>1. Airborne Measurements</u>: Concentrations of the following airborne species were measured in the kiln emissions as well as in the ambient air:
 - Carbon dioxide (CO₂)
 - Carbon monoxide (CO)
 - Methane (CH₄)
 - Total non-methane hydrocarbons (TNMHC)
 - Total suspended particulates (TSP)
 - Nitrous oxide (N₂O)

- <u>2. Solid/liquid Measurements</u>: The following fuel and solid/liquid product parameters were also measured:
 - Wood (starting material): mass (amount used) and contents of moisture, carbon, and energy.
 - Charcoal (product): mass, carbon, and energy content
 - Brands (partly carbonized wood product remaining in chamber) and ash: mass and carbon content
 - TSP: mass and carbon content
 - Condensable liquid emissions (condensables): mass and carbon content
 - Deposited solids: mass

The wood used in these experiments came from trees at the CRC plantation in Saraburi, Thailand. The trees were felled, cut, and the wood was allowed to air dry under a sheltered area of the CRC. For each experiment, wood from this stock was weighed and packed into the main part of the kiln. The weight of the wood used in the firing port of the kiln was also determined. The firing port provides the heat to start the carbonization in the main chamber.

C. Summary of Experimental Procedure (Details in Appendix B)

After pilot runs to polish the protocols, measurements were taken of three successful runs of each kiln type, as shown in Table 1. Thus, the results in this study are based on 15 runs total.

A large hood made of galvanized zinc sheet metal (3 m x 3 m at its base) was suspended about 1 m above the ground over each tested kiln. A heavy plastic curtain was hung around the base of the hood down to the ground. The curtain was not placed in the area directly in front of the kiln so as to not interfere with the kiln operators and to allow for proper circulation of air in and around the kiln. As soon as kiln firing began, a hood blower was turned on along with the sampling pump. The blower drew the kiln emissions through the hood toward the sampling probe via a duct system. The sampling probe was located inside of the duct parallel to the flow. A schematic of the hood and duct system is shown in Figure 1 (also see Figure A-7 in Appendix A).

The sampling configuration consisted of a sampling probe, a TSP sampling cassette holding a quartz fiber filter, a low-flow pump, and a large (80-liter) Tedlar bag. The pump provided a constant sampling flow rate of 100-300 ml/min, depending on the kiln type. If the first large bag was filled before the firing process was completed, additional bags were used until the end of firing process. One 80-liter bag (called the mixing bag) was created at the end of each run by time-weighted filling from the multiple large bags used during continuous emissions sampling. The mixing bag thereby represented a sample integrated over the entire firing process. Small (1-liter) Tedlar bags were filled from the large bags and transported to the laboratory at King Mongkut's Institute of Technology Thonburi (KMUTT) in Bangkok, Thailand for GC analysis. For a subsample, 850-ml stainless-steel canisters were also filled and shipped to the

Oregon Graduate Institute of Science and Technology (OGIST) in Oregon, USA for more detailed analysis. Ambient air samples (large bag and TSP filter) were also collected at the CRC before one run of each kiln type. Appendix C gives details of the analytic procedures used.

Table 1. Sampling Summary

Kiln Expt.	Firing time (h)	Sampling probe	#Large Bags &	Mixing bag used?	Large bag & filter	Each large bag	Ambient bag&filter	Asc canister	# grab samples
- F ·	()	flow rate	Filters	8	replicate	analyzed?	sample	filled?	taken
		(ml/min)	used		taken?	.	taken?		
		,							
BBH-1	40.7	100	4	Y	N	N	Y	N	2
BBH-2	34.0	100	3	Y	Y	Y	Y	Y	5
BBH-3	38.2	100	4	Y	N	N	N	N	3
MBH-1	38.5	100	4	Y	N	N	Y	N	4
MBH-2	41.2	100	4	Y	Y	Y	N	Y	2
MBH-3	60.7	100	5	Y	N	N	N	N	4
SD-1	7	250	2	Y	N	N	Y	N	0
SD-2	4	250	1	N	Y	Y	N	N	0
SD-3	5.5	250	2	Y	N	N	N	N	2
EM-1	21	200	4	Y	N	N	Y	N	0
EM-2	20	200	4	Y	Y	Y	N	Y	0
EM-3	24	200	5	Y	N	N	N	N	3
RHM-1	5	300	1	N	N	Y	Y	N	0
RHM-2	4	300	1	N	Y	Y	N	Y	0
RHM-3	5	300	1	N	N	Y	N	N	2

Large bag - 80-liter Tedlar bag used for continuous emissions sampling

Mixing bag - 80-liter Tedlar bag filled on time-weighted basis from large bags; 1-liter

Tedlar bags were filled from the mixing bags and analyzed at KMUTT

Filter - 37-mm quartz fiber filter used for TSP sampling

Replicate - parallel sampling replicate taken for one integrated sample (80-liter bag and filter)

Canister - 850-ml canister for gas samples; analyzed at OGIST

Asc. - ascarite trap used in filling canisters (for analysis of N_2O)

Grab samples - Samples taken from directly over the top of the kiln into 1-liter Tedlar bags

After the end of the firing, each kiln is sealed and allowed to cool. At this point, charcoal, brands, and ash were removed and weighed. Samples of each were collected and later analyzed for carbon content and calorific value.

Measurements were made of the emissions that deposited onto the hood and inner kiln surfaces in order to determine the total amount of condensable material ultimately emitted from each kiln. Attempts were made in these experiments to measure the amount of condensable

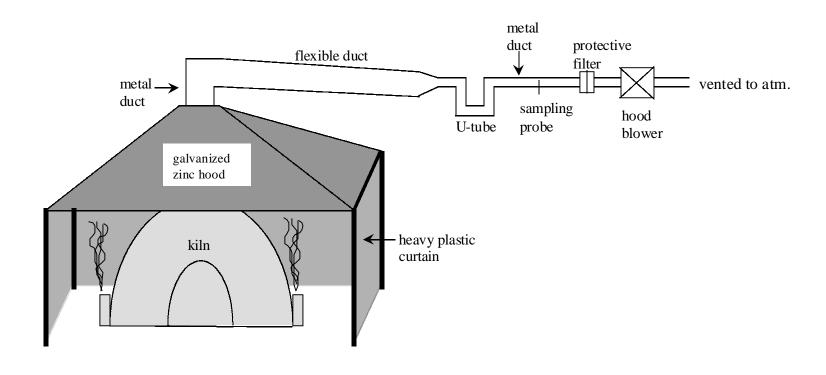


Figure 1. A schematic of the hood and duct system used for emissions sampling in the 15 kiln experiments.

liquids emitted from the kilns by running the air flow through a U-tube. This proved unworkable and, therefore, a set of two follow-up kiln experiments (BBH and EM kilns) were performed in order to better quantify the condensable liquid emissions (condensables). See Appendix D for a detailed description of the follow-up experiments.

A number of activities were performed as part of the quality assurance plan, including sharing of common laboratory standards and cross-laboratory comparisons of split samples. Appendix E provides the details.

IV: RESULTS

A. Airborne Sampling Measurements

The net concentrations of the airborne kiln emission samples were determined by subtracting the ambient concentrations found at the Charcoal Research Centre. The emission sample concentrations for each of the fifteen kiln runs, as determined at KMUTT from the mixing bag samples, are shown in Table 2. The ambient sample concentrations are also shown at the bottom of Table 2 (A-BBH-1, A-BBH-2, etc.). Table 3 shows the net emission ratios to CO_2 on a carbon basis (molar or mass) for CO, CH_4 , TNMHC, TSP, and total products of incomplete combustion (PIC = CO + CH_4 + TNMHC + TSP). The ratios in Table 3, not the net concentrations themselves, are used in the calculation of emission factors for each of the airborne species as shown in section C below.

B. Solid Product Measurements

The total mass and mass of carbon in the solid species involved in each of the fifteen charcoal making experiments (total wood, charcoal, brands, ash, condensables, and TSP) are shown in Table 4. Total wood is the combination of the wood loaded into the kiln (wood in kiln) and wood used for external firing of the kiln (wood for fire). The wood moisture fraction was calculated on the dry basis: moisture fraction = (wet wood mass - oven dry wood mass)/dry wood mass. An average value of 5% moisture content was applied for the charcoal produced. We were not able to determine the total mass of condensable species ultimately emitted from the kilns in the original 15 experiments. Therefore, the emission factor for condensables was determined in a set of two follow-up experiments using BBH and EM kilns. Carbon in the condensable liquid emissions accounted for 2% of the original wood carbon in the follow-up BBH kiln experiment and 4% of that in the follow-up EM experiment, as discussed in Appendix C. An average factor of 3% of the original wood carbon was applied to estimate the amount of condensables carbon emitted in each of the 15 original kiln experiments shown in Table 4.

The mass of emissions that deposited onto the large hood and inner kiln surfaces was determined in each of the 15 experiments. The solids deposited on the inside of the large hood were assumed to be 50% carbon, while those that deposited on inner kiln surfaces were assumed to be 70% carbon (carbon analyses were not performed on these products). The total mass of carbon in these deposited substances ranged from 0.03-0.74% of the original wood carbon for the 15 experiments, with an average of 0.24%. Because of their small contribution to the total carbon outflow from the kilns, these results were not included in Table 4.

Table 5 presents the results of the carbon content and calorific value analyses of the solid species from the 15 original kiln experiments. These carbon content values were used in calculating the carbon masses shown in Table 4. Two batches (labeled "a" and "b") of each of the two wood types were analyzed for carbon content. The charcoal yields, or kiln conversion efficiencies, were determined for each experiment. These data are shown in Table 6. The dry basis charcoal yield is the mass of charcoal produced divided by the total dry mass of wood used in the kiln run (wood inside kiln plus wood for fire). The wet basis charcoal yield is the ratio of

Table 2. Emission sample concentrations (from KMUTT mixing bag data) and ambient concentrations (labeled "A-")

	CO_2	CO	CH_4	TNMHC	TSP	N_2O
	(ppm)	(ppm)	(ppm)	(ppm)	(mg/m^3)	(ppb)
BBH-1	24970	6666	2353	2285	77.8	591
BBH-2	15860	4556	1429	1361	115	572
BBH-3	27770	6102	2212	2980	8.96	809
MBH-1	6437	1422	300	266	4.91	475
MBH-2	5411	981	223	249	11.0	359
MBH-3	7263	1147	328	410	3.54	380
SD-1	10680	3995	1095	1494	66.6	528
SD-2	12800	5150	1410	2568	18.7	NA
SD-3	15570	4200	1456	1793	85.0	NA
EM-1	2060	493	119	718	7.21	341
EM-2	3286	907	162	398	6.22	484
EM-3	3504	737	186	215	12.1	764
RHM-1	10070	1343	225	301	29.7	591
RHM-2	13500	1056	224	153	0.980	968
RHM-3	11930	1376	364	207	4.74	801
A-BBH-1	671	1.12	2.65	5.38	0.825	NA
A-BBH-2	618	0.58	1.63	12.2	NA	NA
A-MBH-1	611	0.35	2.01	NA	NA	NA
A-SD-1	551	NA	7.54	29.4	0.539	NA
A-EM-1	492	NA	NA	24.9	0.480	NA
A-RHM-1	490	NA	NA	12.4	0.170	NA
(KMUTT)						
A-RHM-1	416	1.4	2.0	NA	NA	335
(OGIST)						

NA = not analyzed

carbon produced to the total wet mass of wood used. The carbon yield is the mass of charcoal carbon produced divided by the total mass of carbon in the original wood used. Energy conversion to charcoal is the ratio of the total energy content of the charcoal product to that of the wood input. As expected, the better insulated, more manageable brick kilns (BBH) had the highest average yields, for both charcoal yield and charcoal carbon yield. The earth mound kilns (EM) had the lowest average charcoal carbon yield. Also shown in Table 6 are the brand yields, using both wet and dry mass bases and an energy basis. Brands can be considered a secondary product of charcoal production, because they are often sold as cooking fuel, commanding a price

greater than that of raw wood, but less than that of charcoal. Brands are also often reloaded into a kiln and fully converted to charcoal in a subsequent firing of the kiln.

Table 3. Net emission ratios of gases and TSP to CO2 (ratio of carbon; dimensionless)

	CO/CO_2	CH_4/CO_2	TNMHC/	TSP/CO ₂	PIC/CO ₂ a	$N_2O/CO_2^{\ b}$
			CO_2			
						_
BBH-1	0.2729	0.0962	0.0929	0.00349	0.4655	1.28E-05
BBH-2	0.2975	0.0931	0.0878	0.00824	0.4866	1.50E-05
BBH-3	0.2241	0.0811	0.1089	0.000299	0.4144	2.42E-05
MBH-1	0.2413	0.0504	0.0423	0.000810	0.3349	3.02E-05
MBH-2	0.2016	0.0452	0.0478	0.00230	0.2969	8.02E-06
MBH-3	0.1707	0.0484	0.0586	0.000447	0.2781	1.30E-05
SD-1	0.3943	0.1078	0.1458	0.00535	0.6532	1.77E-05
SD-2	0.4203	0.1148	0.2082	0.00122	0.7446	NA
SD-3	0.2796	0.0968	0.1183	0.00462	0.4993	NA
EM-1	0.3258	0.0764	0.4640	0.00368	0.8698	1.81E-06
EM-2	0.3313	0.0582	0.1393	0.00174	0.5305	3.17E-05
EM-3	0.2491	0.0618	0.0670	0.00324	0.3812	1.30E-04
RHM-1	0.1410	0.0233	0.0298	0.00250	0.1967	3.70E-05
RHM-2	0.0815	0.0171	0.0105	0.0000376	0.1090	6.33E-05
RHM-3	0.1209	0.0317	0.0167	0.000312	0.1696	5.17E-05

NA = not analyzed

^a PIC = products of incomplete combustion = CO + CH₄ + TNMHC + TSP

^b units: molecular ratio

12

Table 4. Solid product measurements

	Wet wood in	Wet wood for	Total wet		Total dry	Total wood C		Charcoal C	Brands	Brands C
	kiln (kg)	fire(kg)	wood (kg)	fraction ^a	wood (kg)	(kg)	(kg)	(kg)	$(kg)^{b}$	(kg)
BBH-1	831.0	42.5	873.5	0.172	745.4	330.0	249.5	192.3	37.0	20.9
BBH-2	755.0	47.0	802.0	0.145	701.4	310.5	238.0	169.9	77.0	46.5
BBH-3	759.0	38.0	797.0	0.182	674.3	294.6	219.0	168.4	57.0	36.7
MBH-1	561.5	40.0	601.5	0.161	518.0	229.3	157.0	114.1	68.0	41.1
MBH-2	575.0	50.0	625.0	0.176	531.3	235.2	171.5	125.0	73.6	44.5
MBH-3	551.0	50.0	601.0	0.171	513.4	224.3	152.8	113.2	34.0	20.6
SD-1	74.5	5.1	78.5	0.267	62.8	27.8	19.0	13.8	1.0	0.59
SD-2	81.0	2.4	83.0	0.204	69.3	30.7	22.4	17.8	1.0	0.59
SD-3	71.0	7.1	77.0	0.183	66.0	28.8	16.9	13.3	0.60	0.35
EM-1	200.0	13.3	211.0	0.205	177.0	78.4	50.0	35.7	6.0	2.9
EM-2	200.0	7.3	206.0	0.215	170.6	75.5	48.5	32.8	31.0	15.4
EM-3	200.0	7.3	206.0	0.210	171.3	74.8	56.0	41.7	30.0	15.2
RHM-1	200.0	8.2	206.5	0.269	164.2	72.7	50.0	39.5	21.0	10.1
RHM-2	200.0	7.4	206.0	0.235	168.0	74.4	40.6	33.5	17.0	8.27
RHM-3	200.0	7.8	206.0	0.299	160.0	69.9	54.9	41.7	12.0	5.91

(continued)

13

SD-2

SD-3

EM-1

EM-2

EM-3

RHM-1

RHM-2

RHM-3

9.9 1.0 0.093 BBH-1 0.254 BBH-2 0.12 9.3 0.469 1.5 BBH-3 1.5 0.15 8.8 0.0170 MBH-1 6.9 1.0 0.061 0.0408 0.20 MBH-2 2.3 7.1 0.104 MBH-3 0.0292 2.5 0.16 6.7 SD-1 1.0 0.14 0.83 0.0401

0.20

0.12

0.17

0.22

0.15

0.56

1.30

0.99

(kg)

Ash C Condensables C

 $(kg)^{c}$

0.92

0.86

2.4

2.3

2.2

2.2

2.2

2.1

Table 4. (continued)

Ash

(kg)

1.5

0.90

3.0

4.5

4.0

8.0

11.0

6.0

 \overline{TSPC} $(kg)^d$

0.00782

0.0438

0.0733

0.0282

0.0365

0.0426

0.000986 0.00512

^a Determined on a dry basis; moisture fraction = (wet mass-dry mass)/dry mass

^b The mass of brands produced in the BBH-3 run was set as the average of the masses of brands produced in the other two BBH runs

^c Set equal to 3.0% of wood C (as determined in the BBH and EM follow-up experiments)

^d TSP was determined from its ratio to CO₂ in the airborne emissions samples

Table 5. Carbon analyses and calorific values of solid products

	Charcoal	Charcoal	Brands	Brands	Ash	TSP
	% carbon	calorific	% carbon	calorific	% carbon	% carbon
		value (kJ/g)		value(kJ/g)		
BBH-1	80.97	29.70	59.28	23.07	8.83	NA
BBH-2	75.00	28.38	NA	NA	7.87	54.1
BBH-3	80.79	30.57	67.72	26.54	10.17	46.7
MBH-1	76.34	28.77	NA	NA	6.11	NA
MBH-2	76.59	29.17	NA	NA	8.91	51.7
MBH-3	77.81	29.74	NA	NA	6.58	NA
SD-1	76.52	NA	61.49	24.02	13.67	NA
SD-2	83.47	30.59	NA	NA	NA	40.2
SD-3	82.50	30.14	61.41	23.93	12.96	NA
EM-1	75.00	NA	51.48	NA	5.82	NA
EM-2	71.02	NA	NA	NA	NA	40.0
EM-3	78.24	NA	53.18	NA	3.86	NA
RHM-1	83.00	NA	50.43	NA	7.03	NA
RHM-2	86.60	NA	NA	NA	NA	NA
RHM-3	79.74	NA	51.74	NA	16.52	NA
Wood Samples	Wood	Wood				
•	% carbon	calorific				
		value (kJ/g)				
		, <u>0</u> ,				
Eucalyptus (a)	44.52	20.41				
Eucalyptus (b)	44.02	19.82				
Leucaena (a)	43.67	18.88				
Leucaena (b)	43.70	19.62				
NIA	1					

NA = not analyzed

15

Table 6. Charcoal and brands yields (mass, carbon, and energy bases)

	Charcoal yield (wet basis) ^a	Charcoal yield (dry basis) b	Charcoal carbon yield ^c	Energy conversion to charcoal	Brands yield (wet basis) ^a	Brands yield (dry basis) b	Brands carbon yield ^c	Brands conversion to charcoal
	(wei busis)	(ary vasis)	carbon yieia	to charcoat	(wei basis)	(ary basis)	унена	to charcoat
BBH-1	0.286	0.335	0.583	0.471	0.0424	0.0496	0.0633	0.0542
BBH-2	0.297	0.339	0.547	0.456	0.0960	0.110	0.150	
BBH-3	0.275	0.325	0.572	0.491	0.0715	0.0845	0.125	0.111
MBH-1	0.261	0.303	0.498	0.413	0.113	0.131	0.179	
MBH-2	0.274	0.323	0.532	0.446	0.118	0.139	0.189	
MBH-3	0.254	0.298	0.505	0.438	0.0566	0.0662	0.0916	
SD-1	0.242	0.303	0.498		0.0127	0.0159	0.0211	0.0181
SD-2	0.270	0.323	0.580	0.468	0.0120	0.0144	0.0191	
SD-3	0.219	0.256	0.460	0.382	0.00779	0.00909	0.0122	0.0108
EM-1	0.237	0.282	0.456		0.0284	0.0339	0.0375	
EM-2	0.235	0.284	0.434		0.150	0.182	0.204	
EM-3	0.272	0.327	0.557		0.146	0.175	0.203	
RHM-1	0.242	0.305	0.544		0.102	0.128	0.139	
RHM-2	0.197	0.242	0.450		0.0825	0.101	0.111	
RHM-3	0.267	0.343	0.596		0.0583	0.0750	0.0846	

a wet basis yield = product mass/wet wood mass
b dry basis yield = product mass / dry wood mass
c carbon yield = product carbon mass / wood carbon mass

C. Carbon Balance and Calculation of Emission Factors

Starting with a carbon balance on the system, we calculated the emission factors for each of the species of interest (CO_2 , CO, CH_4 , TNMHC, TSP, N_2O). We will present the emission factors in four different forms, each having its own utility depending on the desired end use:

- a. grams of pollutant emitted per kilogram of dry wood loaded into the charcoal kiln
- b. grams of pollutant emitted per kg of charcoal produced
- c. grams of pollutant carbon emitted per kg of wood carbon
- d. grams of pollutant carbon emitted per kg of charcoal carbon produced.

The carbon balance for the charcoal making process can be written as follows, on a carbon basis:

wood = charcoal + brands + ash + condensable liquids + CO₂+ CO + CH₄ + TNMHC + TSP,

dividing through by CO₂ and rearranging yields:

$$1 = (wood - charcoal - brands - ash - condensables)/CO2 - (CO + CH4 + TNMHC + TSP)/CO2.$$

We define $(CO + CH_4 + TNMHC + TSP)/CO_2 = K$.

The total amount of CO₂ emitted (still in terms of carbon) was found by solving the above equation:

$$CO_2 = (wood - charcoal - brands - ash - condensables)/(1 + K)$$

The absolute CO_2 emission factor was then found by dividing this total amount of CO_2 emitted by the total amount of wood used in the kiln run or charcoal produced in the process. The emission factors for the other species of interest were found via their emission ratios to CO_2 (again on a carbon basis). All four forms of the emission factors for all airborne species were calculated, and the average values for the three runs per kiln are shown in Table 7a-d. The coefficients of variation were calculated for the first set of emission factors and are shown in the lower half of Table 7a.

The distribution of wood carbon for each of the runs and the means for each of the five kiln types is shown in Table 8. Figure 2 shows the balance for the averages for each of the five kilns. They range from the best performance in BBH in which only 290 g C is diverted to PIC and CO₂ to the worst, SD, in which 435 g is lost to airborne species.

Table 7a. Average emission factors, grams of pollutant per kilogram charcoal produced, and coefficients of variation (CV)

	CO_2	CO	CH_4	$TNMHC^a$	TSP	PIC	gases+TSP	N_2O
brick beehive (BBH)	966	162	31.8	29.7	1.90	226	1192	0.0166
mud beehive (MBH)	1235	158	21.7	19.9	0.693	200	1436	0.0212
single drum (SD)	1517	336	57.7	71.5	4.19	470	1987	0.0259
earth mound (EM)	1140	226	27.7	95.3	2.25	351	1491	0.0458
rice husk mound (RHM)	1570	106	12.7	8.53	0.807	128	1699	0.0843
	Coefficien	ts of varia	tion (CV)	for above av	verage em	ission fac	tors	
ввн	0.10	0.16	0.16	0.15	0.92	0.13	0.10	0.27
MBH	0.25	0.19	0.27	0.41	0.60	0.20	0.24	1.43
SD	0.34	0.15	0.25	0.08	0.70	0.14	0.29	0.23
EM	0.32	0.43	0.44	1.11	0.55	0.59	0.38	0.99
RHM	0.44	0.19	0.16	0.36	1.42	0.17	0.42	0.19

Table 7b. Average emission factors, grams of pollutant per kilogram dry wood

	CO_2	CO	CH_4	$TNMHC^a$	TSP	PIC	gases+TSP	N_2O
brick beehive (BBH)	322	54.2	10.6	9.88	0.640	75.3	397	0.00551
mud beehive (MBH)	378	48.4	6.63	6.08	0.217	61.4	439	0.00645
single drum (SD)	434	98.1	16.6	20.9	1.17	136.8	571	0.00775
earth mound (EM)	334	65.7	8.09	27.2	0.660	101.6	435	0.0144
rice husk mound (RHM)	443	30.9	3.71	2.51	0.248	37.4	480	0.0232

Table 7c. Average emission factors, grams of pollutant C per kilogram charcoal C produced

	CO_2	CO	CH_4	TNMHC	TSP	PIC	gases+TSP	N_2O^b
brick beehive (BBH)	350	92.9	31.7	33.8	0.380	159	509	0.0120
mud beehive (MBH)	460	92.6	22.2	23.2	0.131	138	598	0.0158
single drum (SD)	539	188.3	56.3	79.5	0.604	325	864	0.0192
earth mound (EM)	441	137.6	29.3	115	0.345	282	723	0.0345
rice husk mound (RHM)	536	57.4	12.0	9.21	0.112	78.7	615	0.0572

Table 7d. Average emission factors, grams of pollutant C per kilogram wood C

	CO_2	СО	CH_4	TNMHC	TSP	PIC	gases+TSP	N_2O^c
brick beehive (BBH)	199	52.7	18.0	19.2	0.779	90.6	290	0.00683
mud beehive (MBH)	234	47.1	11.3	11.8	0.249	70.5	305	0.00798
single drum (SD)	269	95.4	28.3	40.6	1.07	165	435	0.00318
earth mound (EM)	206	63.8	13.7	52.6	0.599	131	337	0.0179
rice husk mound (RHM)	274	30.0	6.31	4.86	0.224	41.4	315	0.0287

 $PIC = products of incomplete combustion = CO + CH_4 + THMHC + TSP$

gases+TSP = $CO_2 + CO + CH_4 + THMHC + TSP$ ^a Assuming a per carbon molecular weight of 14
^b In units of N atoms per 1000 atoms of charcoal C
^c In units of N atoms per 1000 atoms of wood C

Table 8. Percent distribution of the original wood carbon in the products of the charcoal making process

	Charcoal	Brands	Condensables	CO_2	CO	CH_4	TNMHC	Ash	TSP
			•				• • •	0.0001	
BBH-1	58.3	6.33	3.0	22.1	6.03	2.12	2.05	0.0281	0.0770
BBH-2	54.7	15.0	3.0	18.3	5.45	1.71	1.61	0.0380	0.151
BBH-3	57.2	12.5	3.0	19.3	4.33	1.57	2.10	0.0518	0.00578
MBH-1	49.8	17.9	3.0	21.9	5.29	1.11	0.929	0.0266	0.0178
MBH-2	53.2	18.9	3.0	19.1	3.86	0.866	0.914	0.0871	0.0440
MBH-3	50.5	9.16	3.0	29.2	4.98	1.41	1.71	0.0733	0.0130
SD-1	49.8	2.11	3.0	27.0	10.6	2.91	3.93	0.492	0.144
SD-2	58.0	1.91	3.0	20.9	8.77	2.40	4.35	0.650	0.0255
SD-3	46.0	1.22	3.0	32.9	9.21	3.19	3.89	0.404	0.152
EM-1	45.6	3.75	3.0	25.4	8.27	1.94	11.8	0.223	0.0935
EM-2	43.4	20.4	3.0	21.5	7.12	1.25	2.99	0.288	0.0374
EM-3	55.7	20.3	3.0	15.0	3.74	0.929	1.01	0.206	0.0487
RHM-1	54.4	13.9	3.0	23.4	3.30	0.546	0.698	0.773	0.0586
RHM-2	45.0	11.1	3.0	35.3	2.87	0.602	0.370	1.75	0.00133
RHM-3	59.6	8.46	3.0	23.5	2.84	0.745	0.392	1.42	0.00733
Averages:									
brick beehive	56.7	11.3	3.0	19.9	5.27	1.80	1.92	0.0393	0.0779
mud beehive	51.1	15.3	3.0	23.4	4.71	1.13	1.18	0.0623	0.0240
single drum	51.3	1.74	3.0	26.9	9.54	2.83	4.06	0.516	0.107
earth mound	48.2	14.8	3.0	20.6	6.38	1.37	5.26	0.239	0.0599
rice husk mound	53.0	11.2	3.0	27.4	3.00	0.631	0.486	1.31	0.0224

Note: any differences between averages of the displayed sample values and the displayed averages are due to rounding.

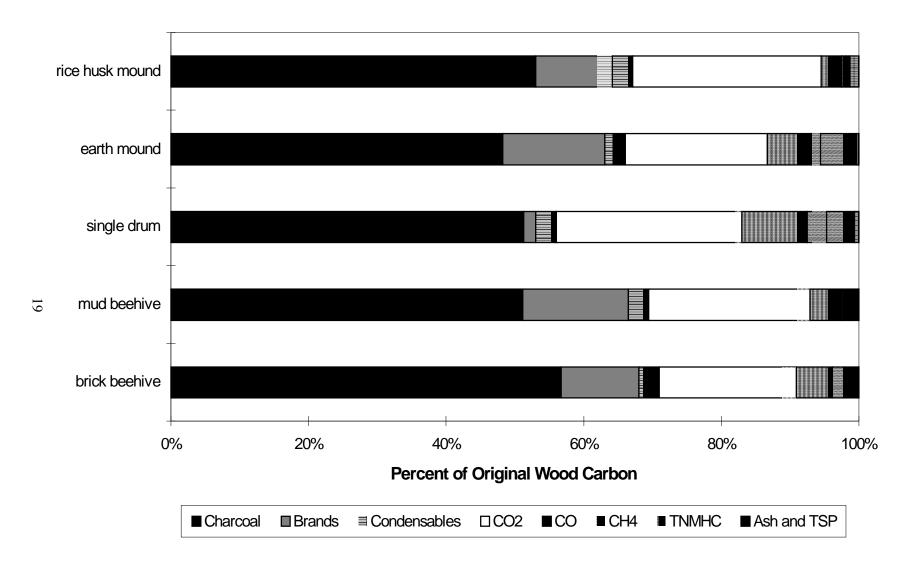


Figure 2. Distribution of the original wood carbon in the products of the charcoal-making process.

D. GC Analysis of Canister Samples

The data on gaseous emissions generated from the analyses of the canister samples (from each of the 15 kiln experiments) at OGIST (as a part of the Quality Assurance Plan - Appendix E) were treated in the same fashion as the gaseous emissions data generated from analyses of the mixing bag samples at KMUTT. Emission ratios of CO, CH₄, TNMOC (total non-methane organic compounds), and the sum of those three species to CO₂ were determined using the OGIST GC data. These emission ratios were compared to the ratios determined from the KMUTT data set (Table 3), as shown in Table 9. The percent difference between the two values is given, using the KMUTT data as the denominator. Please note that KMUTT measured total non-methane hydrocarbons (TNMHC), while OGIST measured total non-methane organic compounds (TNMOC). Although it is not possible to make an exact comparison between TNMHC and TNMOC due to the difference in the analytical techniques, we did display these numbers, because the comparison is still of some value. This difference in the analytical techniques explains the greater variability (higher percent differences) between KMUTT and OGIST in this case compared to the inter-laboratory differences seen in the cases of CO₂, CO, and CH₄, as seen in Table 9.

For comparison, the emission ratios from the OGIST data were plotted against those from the KMUTT data, shown in Figures 3a-c along with the slope and correlation coefficient (r²). Note that the r² ranged from 0.91 (methane) to 0.44 (hydrocarbons).

The OGIST emission ratios were also used to calculate the emission factors (in units of grams of pollutant per kilogram wood) for each of the 15 experiments. A comparison of the OGIST-data emission factors with the earlier calculated KMUTT-data emission factors (from Table 7) is shown in Table 10. Note that because of the insensitivity of the carbon balance method to variations in individual concentration measurements, most differences in emission factors are relatively small, except for TNMHC where for some single runs, they differed by a factor of two or more. In the first Earth Mound run, the difference reached nearly a factor of six.

E. Grab Samples for Determining Emission Ratios and Emission Factors

Since building of a large hood for continuous monitoring would be impractical for kilns in remote locations, we wished to test how well grab sampling above the chimneys of the kilns would replicate continuous sampling in the hood. From 2 to 5 grab samples were taken during each of 9 kiln runs. These grab samples were analyzed at KMUTT along with the mixing bag samples (primary method). Time-weighted averages of the net emission concentrations found for each of the grab samples for a given kiln run were calculated. These grab sample average values were used to calculate emission ratios and, subsequently, emission factors. Comparisons of the emission ratios and absolute emission factors determined from the grab sample data to those determined from the mixing bag data (from Tables 3 and 7) are shown in Tables 11 and 12. The percentage differences are also given, using the mixing bag method as the denominator and are highest for hydrocarbons. Note that runs where only 2 grab samples were taken account for the majority of the instances where the percent differences were the largest (e.g., MBH-2 where there was a factor of three difference for hydrocarbons). More grab samples allow for better

characterization of the true gaseous emissions. Although the grab samples do not exactly duplicate what was found in the mean hood samples, the variation is similar to that among hood runs.

22

Table 9. Comparison of emission ratios of CO, CH₄, and TNMHC to CO₂ determined using OGIST canister data and KMUTT mixing bag data

	CO/CO_2				CH_4/CO_2			TNMHC/C	CO_2	(CO+	CH_4+TNN	$MHC)/CO_2$
Sample name			%			%			%			%
	KMUTT	OGIST	difference	KMUTT	OGIST	difference	KMUTT	OGIST ^a	difference	KMUTT	OGIST	difference
BBH-1M	0.267	0.252	-5.7	0.0942	0.0934	-0.90	0.0343	0.0285	-15	0.395	0.374	-5.3
BBH-2M	0.287	NR		0.0901	NR		0.0312	0.0387	24	0.408	NR	
BBH-2B-R1	0.321	0.296	-7.9	0.107	0.0687	-36	0.0559	0.0509	-8.8	0.484	0.415	-14
BBH-3M	0.220	NR		0.0796	NR		0.0390	0.0467	20	0.338	NR	
MBH-1M	0.221	0.237	7.3	0.0466	0.0465	-0.20	0.0147	0.0298	103	0.282	0.313	11
MBH-2M	0.181	0.230	27	0.0412	0.0493	20	0.0164	0.0293	79	0.239	0.308	29
MBH-2B-R1	0.187	0.241	29	0.0332	0.0400	21	0.0217	0.0222	2.4	0.242	0.303	25
MBH-3M	0.158	0.243	54	0.0452	0.0624	38	0.0207	0.0347	68	0.224	0.340	52
SD-1M	0.374	0.206	-45	0.103	0.0653	-36	0.0510	0.0284	-44	0.528	0.299	-43
SD-3M	0.270	NR		0.0935	NR		0.0418	0.0509	22	0.405	NR	
EM-1M	0.239	0.211	-12	0.0578	0.0673	17	0.127	0.0192	-85	0.424	0.298	-30
EM-2M	0.276	0.176	-36	0.0493	0.0422	-14	0.0440	0.0130	-70	0.369	0.231	-37
EM-2-R2	0.305	0.252	-18	0.0654	0.0665	1.6	0.0253	0.0296	17	0.396	0.348	-12
EM-3M	0.210	0.145	-31	0.0531	0.0449	-15	0.0222	0.0194	-12	0.286	0.210	-27
A-RHM-1	0.000	0.00337		0.000	0.00481		0.00918	0.00130	-86	0.00918	0.00948	3.2
RHM-1M	0.133	0.105	-21	0.0224	0.0234	4.7	0.0110	0.0135	22	0.167	0.142	-15
RHM-2M	0.0782	0.0889	14	0.0166	0.0197	19	0.00412	0.00880	114	0.0990	0.117	19
RHM-3M	0.115	0.116	0.17	0.0305	0.0255	-17	0.00652	0.0149	129	0.152	0.156	2.3

Note: any differences between percent differences of the displayed values and the displayed percent differences are due to rounding. NR = not reported

^a OGIST measured TNMOC (total non-methane organic compounds), while KMUTT measured TNMHC (total non-methane hydrocarbons).

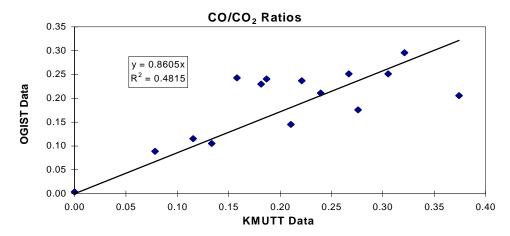


Figure 3a. Comparison of CO/CO₂ emission ratios from KMUTT data and OGIST data

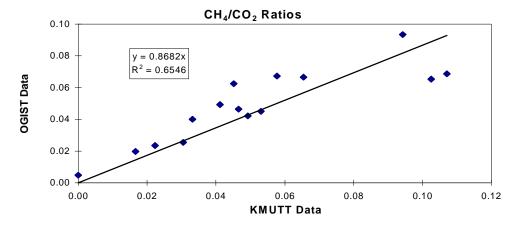


Figure 3b. Comparison of CH₄/CO₂ emission ratios from KMUTT data and OGIST data

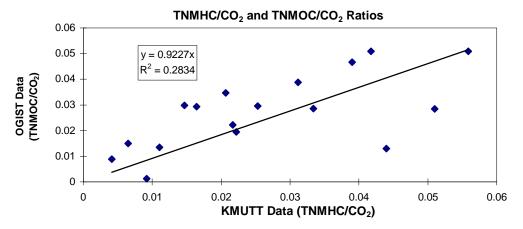


Figure 3c. Comparison of TNMHC/CO $_2$ emission ratios from KMUTT data to TNMOC/CO $_2$ ratios from OGIST data

Table 10. Comparison of emission factors determined using OGIST canister GC data and KMUTT mixing bag GC data (g pollutant/kg dry wood)

	CO_2		C	0	C	H_4	TNM	HC^{a}	T_{s}	SP^{b}	P	PIC	gases	es+TSP	
		KMUTT		KMUTT		KMUTT		KMUTT		KMUTT		KMUTT		KMUTT	
	data	data	data	data	data	data	data	data	data	data	data	data	data	data	
BBH-1	365	358	59.7	62.2	12.7	12.6	9.31	10.6	0.784	0.631	82.5	86.0	448	444	
MBH-1	340	356	55.8	54.7	6.22	6.54	9.64	4.80	0.185	0.152	71.9	66.2	412	422	
MBH-2	285	311	47.4	39.9	5.77	5.12	8.30	4.73	0.563	0.377	62.1	50.1	347	361	
MBH-3	412	467	71.4	50.8	10.4	8.23	14.0	8.72	0.211	0.122	96.0	67.8	508	535	
SD-1	529	438	71.9	110	13.0	17.2	13.7	20.3	1.78	1.59	100.4	149.0	630	587	
EM-1	561	412	84.7	85.4	15.3	11.5	10.5	60.9	0.636	1.03	111.2	158.8	672	571	
EM-2	418	348	50.9	73.5	6.92	7.38	5.13	15.5	0.288	0.413	63.2	96.7	481	445	
EM-3	261	241	27.1	38.2	4.75	5.42	4.95	5.14	0.515	0.532	37.3	49.2	298	290	
RHM-1	385	379	27.2	34.1	3.44	3.22	4.84	3.61	0.904	0.648	36.4	41.5	422	421	
RHM-2	558	572	32.8	29.7	4.13	3.56	4.45	1.91	0.0185	0.0147	41.4	35.2	599	608	
RHM-3	370	376	28.4	28.9	3.56	4.35	5.03	2.00	0.0993	0.0800	37.1	35.4	407	412	

^a Assuming a per carbon molecular weight of 14
^b TSP was not determined at OGIST. The "OGIST" TSP emission factors were determined using the KMUTT TSP data along with CO₂ data from OGIST canister analyses.

25

Table 11. Comparison of grab sample method to mixing bag method: emission ratios of CO, CH_4 , and TNMHC to CO_2

	# of	CO	O/CO_2			CH_4/CO_2 TNMHC/CO ₂				$(CO+CH_4+TNMHC)/CO_2$			
	grabs taken	grab	mixing bag	% difference	grab	mixing bag	% difference	U	mixing bag	% difference	grab	mixing bag	% difference
				3,5			35			33			35
BBH-1	2	0.302	0.273	11	0.125	0.0963	30	0.121	0.0930	30	0.549	0.463	19
BBH-2	5	0.317	0.298	6.6	0.123	0.0933	32	0.150	0.0889	69	0.591	0.480	23
BBH-3	3	0.231	0.224	2.9	0.134	0.0812	65	0.0837	0.109	-23	0.449	0.415	8.3
MBH-1	4	0.259	0.242	7.1	0.0521	0.0508	2.7	0.0645	0.0452	43	0.376	0.338	11
MBH-2	2	0.306	0.203	51	0.0770	0.0456	69	0.242	0.0513	371	0.624	0.300	108
MBH-3	4	0.120	0.171	-30	0.0303	0.0487	-38	0.0301	0.0597	-50	0.180	0.280	-36
SD-3	2	0.356	0.280	27	0.153	0.0970	58	0.262	0.119	121	0.771	0.496	56
EM-3	3	0.288	0.251	14	0.0778	0.0627	24	0.0700	0.0685	2.2	0.435	0.382	14
RHM-3	2	0.0822	0.121	-32	0.0129	0.0318	-59	0.0111	0.0170	-35	0.106	0.170	-37

Note: any differences between percent differences of the displayed values and the displayed percent differences are due to rounding.

26

Table 12. Comparison of emission factors determined using grab sample method and mixing bag method (g pollutant/kg dry wood)

	CO_2			(CO			CH_4		,	TNMHC ^a	MHC ^a			
	grab method	mixing bag	% difference	grab method	mixing bag	% difference	grab method	mixing bag	% difference	grab method	mixing bag	% difference			
BBH-1	338	358	-5.5	65.1	62.2	4.6	15.4	12.6	23	13.1	10.6	24			
ВВН-2	276	297	-7.0	55.9	56.3	-0.57	12.4	10.1	23	13.2	8.32	59			
ВВН-3	302	309	-2.4	44.3	44.1	0.53	14.8	9.13	62	8.05	10.7	-25			
MBH-1	345	356	-3.1	57.0	54.7	4.2	6.55	6.54	0.24	7.10	4.80	48			
MBH-2	247	311	-20	48.2	39.9	21	6.94	5.12	36	19.1	4.73	303			
MBH-3	506	467	8.3	38.5	50.8	-24	5.59	8.23	-32	4.85	8.72	-44			
SD-3	445	527	-16	101	93.8	7.3	24.8	18.6	34	37.2	19.9	87			
EM-3	231	241	-4.1	42.2	38.2	11	6.54	5.42	21	5.15	5.14	0.24			
RHM-3	398	376	5.7	20.8	28.9	-28	1.87	4.35	-57	1.40	2.00	-30			

(continued)

Table 12. (continued)

	7	ΓSP			PIC		gases+TSP			
	grab method	mixing bag	% difference	grab method	mixing bag	% difference	grab method	mixing bag	% difference	
BBH-1	0.450	0.631	-29	94.0	86.0	9.3	432	444	-2.7	
BBH-2	1.11	1.24	-10	82.6	75.9	8.8	359	373	-3.8	
BBH-3	0.0548	0.0541	1.4	67.2	64.0	5.0	369	373	-1.1	
MBH-1	0.148	0.152	-3.1	70.8	66.2	7.0	416	422	-1.5	
MBH-2	0.499	0.377	32	74.6	50.1	49	322	361	-11	
MBH-3	0.0731	0.122	-40	49.0	67.8	-28	555	535	3.7	
SD-3	1.88	1.65	14	164	134	23	609	661	-7.8	
EM-3	0.755	0.532	42	54.7	49.2	11	285	290	-1.5	
RHM-3	0.0814	0.0800	1.7	24.2	35.4	-32	422	412	2.5	

Note: any differences between percent differences of the displayed values and the displayed percent differences are due to rounding.

^a Assuming a per carbon molecular weight of 14

V: DISCUSSION

Table 13 presents a summary of earlier charcoal kiln emission studies. The study by Brocard et al. utilized a sampling probe located directly inside of the tested kiln (for some of the samples), and, hence, the results may not so accurately reflect what is ultimately emitted from the kiln. Note that Missouri kilns (USEPA, 1995) are industrial kilns much larger (17.6 tons or more of charcoal produced per cycle) than those commonly used in developing nations, where almost all of the world's charcoal is produced.

As shown in Table 13, the results of this study roughly validate default values listed by IPCC for charcoal kilns in that the IPCC values (IPCC, 1997) fall within the range of the kilns monitored here.¹ The IPCC values are closest to those of the brick beehive kiln in Thailand. Depending on the kiln type, however, compared to the IPCC default values, our values range from -2x to + 1.6x for CO; -1.4x to +1.9x for CH₄; and -6.0x to +1.9x for TNMOC. Thus, to accurately reflect actual emissions, there is need to specify the fraction of charcoal produced in each major kiln type.

One way of looking at the greenhouse-gas implications of charcoal-making can be seen in Figures 4 and 5 for the two most extreme kilns tested, BBH and SD, respectively. Here, two types of global warming commitments (GWC) are applied. The first, called "primary GWC" assumes GWPs only for CO₂ CH₄, and N₂O (respectively, 1, 23, and 290 by mole - IPCC, 1995). The second, "total GWC," applies GWPs also for CO and hydrocarbons, which are less certain (respectively, 4.5 and 12 - IPCC, 1990). For each, the GWC is calculated under two extreme assumptions: that the kilns rely on a completely renewable wood supply, i.e. all carbon is recycled back into trees, compared to the case in which there is complete deforestation, i.e., no carbon recycling to biomass. In the latter case, of course, the GWC includes the full complement from CO₂ (GWP=1.0). In the former, no contribution from CO₂ is included.

We estimated the amount of charcoal produced in each type of kiln in Thailand. The amounts were based upon data on charcoal consumption in Thailand in 1996 (Thailand Department of Energy Development and Promotion, 1996). The fractional use of each kiln type throughout Thailand is shown in the left side of Table 14a-b. Note that sawdust mounds are kilns used to produce charcoal with wood waste from sawmills. Production of charcoal from sawdust mounds has risen rapidly in Thailand during the past several years. Although we did not perform tests on sawdust mound kilns, we believe their emissions patterns to be similar to the rice husk mound because of their similarity of design. Using the annual amount of charcoal produced in Thailand, the fractional use pattern, and the experimental emission factors determined in this study, the total amounts of greenhouse gases emitted from charcoal production in Thailand were calculated. This is shown in Table 14a-b as megatons of pollutant (Mt) and as megatons of pollutant carbon (MtC).

28

¹ Oddly, however, the default charcoal conversion factor used by IPCC, 21%, is substantially lower than we observed. This may be due to its use of a different definition of "conversion efficiency," which the IPCC document unfortunately does not explain.

Finally, using 20-year global warming potentials (GWPs), the total global warming commitments (GWCs) for Thailand's and the world's charcoal kilns are estimated in Table 15. This makes the extreme assumption that the mean emissions for Thai kilns can reasonably be applied to all the world's kilns. Shown for both Thailand and the world is a two-by-two matrix of GWCs, divided as discussed above for Figures 4 and 5. The global GWC numbers are based on the assumption that all of the charcoal produced globally comes from the following mix of kiln types: 50% EM (roughly representing the mobile informal-sector² kilns of Africa), 25% BBH (roughly representing the large commercial kilns of Brazil), and 25% from the mix of kilns as in Thailand (roughly representing Asia). Also shown for comparison are the total estimated fossil-fuel contributions to the GWC of Thailand and the world. To pinpoint these values more precisely, however, we are now proceeding to measure emissions from African and Brazilian kilns.

A full analysis of the charcoal fuel cycle for comparison with other fuel cycles would not only require evaluation of the final end use, e.g., cookstoves, as well as the kilns, but also the alternate fate of the wood input if it were not used for charcoal making. For example, in some locations part of the fuel may decay anaerobically in the environment leading to considerable CH₄ emissions. In such a case, the accounting should allocate only the net change in emissions to the charcoal fuel cycle.

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² An informal-sector charcoal kiln is one that is not registered with the authorities, usually made of temporary materials, and is often constructed by one or a few households only in those times of year when agricultural work demand is low.

30

Table 13. Summary of previous charcoal-making kiln emission studies

Study	Kiln Type	% Charcoal yield		En	ission fact	ors (g of pol	lutant per	kg of chard	oal produ	ced)	
		(charcoal mass/ dry wood mass)	CO_2	СО	CH_4	TNMHC	N_2O	NO_x	TSP	Condensables (tars and oils)	Acids
present study	brick beehive	33.3	966	162	31.8	29.7	0.0166		1.90	58	
present study	mud beehive	30.8	1235	158	21.7	19.9	0.0212		0.693	63	
present study	single drum	29.4	1517	336	57.7	71.5	0.0259		4.19	66	
present study	earth mound	29.8	1140	226	27.7	95.3	0.0458		2.25	65	
present study	rice husk mound	29.7	1570	106	12.7	8.53	0.0843		0.807	65	
Brocard et al. (1996)	African earth mound	27.6	1593	254	39	7.2 (as C)	0.11	0.24	14 (as C)		0.72 (as C) (a)
EPA AP-42 (1995)	Missouri		550	145	55	80 (b)		12	(c)	155 (c)	
IPCC Reference Manual (1997)	(world average)	20.8 (d)		210	30	51		0.3			
Shah et al. (1992)	metal partial- combustion kiln	32.7	1192	336	(e)	72 (f)				133 (g)	71

⁽a): sum of formic and acetic acids only

⁽b): derived by subtracting CH₄ from the emission factor given for volatile organic compounds (VOC)

⁽c): TSP was included in the condensables

⁽d): Converted to a dry basis by assuming 20% moisture in wood.

⁽e): CH₄ included in the TNMHC column

⁽f): sum of CH₄, ethane, and ethene

⁽g): sum of tars, phenols, and furfurals

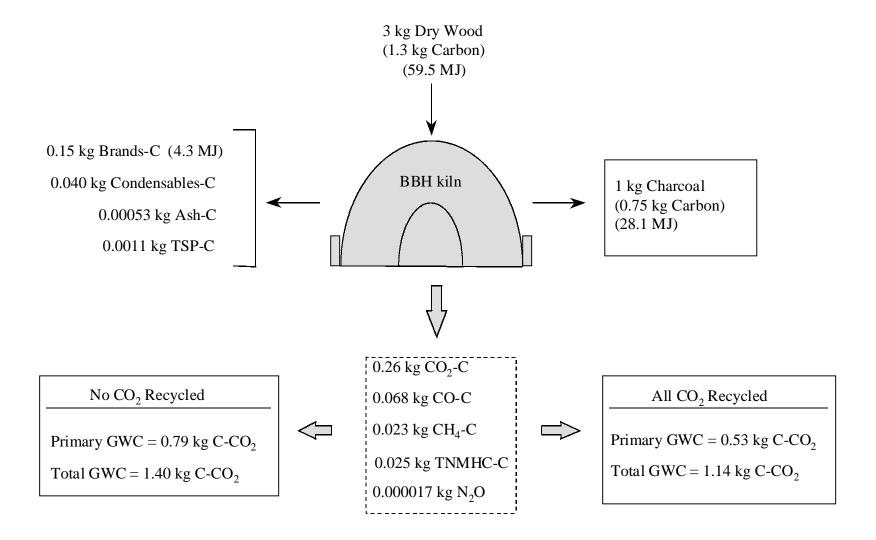


Figure 4. Carbon cycle for charcoal making in the BBH kiln and the GWC resulting from renewable and non-renewable harvesting of the wood.

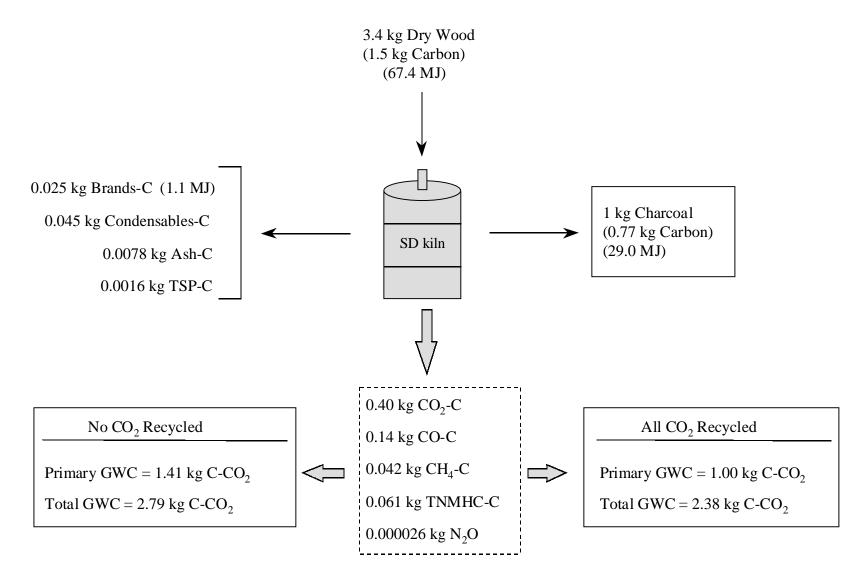


Figure 5. Carbon cycle for charcoal making in the SD kiln and the GWC resulting from renewable and non-renewable harvesting of the wood.

Table 14a. Estimated annual air pollution emissions from charcoal production in Thailand, 1996 a

Kiln type	fractional use in Thailand	CO_2 (Mt)	CO (Mt)	CH_4 (Mt)	$TNMHC (Mt)^b$	TSP (Mt)	N_2O (Mt)
BBH and MBH	0.26	2.1	0.30	0.050	0.047	0.0024	0.000036
EM and RHM	0.21	2.0	0.25	0.030	0.078	0.0023	0.000098
Sawdust Mound	0.53	6.0	0.41	0.048	0.033	0.0031	0.00032
TOTAL	1.0	10.1	0.96	0.129	0.157	0.0078	0.00046

Table 14b. Estimated annual air pollution emissions (as carbon) from charcoal production in Thailand (1996) ^a

Kiln type	fractional use	CO_2	CO	CH_4	TNMHC	TSP	N_2O
	in Thailand	(MtC)	(MtC)	(MtC)	(MtC)	(MtC)	(MtN)
BBH and MBH	0.26	0.56	0.13	0.038	0.040	0.0012	0.000023
EM and RHM	0.21	0.56	0.11	0.023	0.067	0.00092	0.000062
Sawdust Mound	0.53	1.6	0.17	0.036	0.028	0.0012	0.00021
TOTAL	1.0	2.8	0.41	0.097	0.135	0.0034	0.00029

Note: any differences between summations of the displayed values and the displayed TOTAL values are due to rounding.

^a Using a total production of charcoal in Thailand of 7.2 Mt

^b Assuming a per carbon molecular weight of 14

Table 15. Estimated global warming commitments (GWC) from Thai and world charcoal production compared to GWC from fossil fuel cycle

	Thailand primary GWC	Thailand total GWC	World Primary GWC ^a	World total GWC ^a
		(MtC as C	$(O_2 \ equivalents)^b$	
CO ₂ recycled	2.26	5.72	11.8	37.4
CO ₂ not recycled	5.02	8.49	20.1	45.6
GWC from fossil fuel use ^c	67.4	78.9	7887	9749

^a Assuming global charcoal is produced from the following mix of kiln types: 50% EM (Africa), 25% BBH (Brazil), and 25% from a mix of kilns as in Thailand (Asia) ^b Using 20-year global warming potentials ^c For Thailand, primary GWC includes CO₂ and CH₄ only and total GWC includes CO₂, CH₄,

and CO only

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Appendix A: Construction and Operation of the Charcoal Kilns

All kilns were constructed and operated by the CRC staff, which has had long experience with these models and wood species.

Brick Beehive Kiln (BBH)

- Construction: The brick beehive kiln used was a typical commercial charcoal kiln, but slightly smaller in size. The raw materials for constructing the BBH kiln were bricks, clay, and sand. An area of 4 m by 4 m was cleared for the kiln. The foundation was dug out to a depth of three layers of brick. When three brick layers of foundation were constructed, spaces for four chimneys, a loading port, and a firing port were left before the base wall was begun. The brick arrangement used for the walls from the foundation to the top of the kiln was the same as in normal brick construction. The cementing material was made from soft clay and sand. A space for an accelerator hole was left in the wall at 0.8 m above the kiln floor opposite the space for the firing port. Four chimneys were constructed after completion of the kiln wall. Drying and curing of the binding materials was carried out by burning 1-2 armfuls of firewood inside the kiln for 3-4 hours. A diagram and a picture of the BBH kiln are given in Figures A-1 and A-2, respectively.
- Operation: The kiln was loaded by vertically stacking smaller pieces of wood on the kiln floor and placing larger pieces above the smaller pieces and near the firing port. The wood was stacked as close as possible in order to produce more charcoal per kiln run. Areas adjacent to the chimney outlet holes were left clear of wood for at least a 10 cm by 10 cm area. The wood was loosely packed near the firing port. The kiln was lit by combusting firewood in the firing port. The kiln temperature was increased slowly after firing via the hot air from the combusting wood in the firing port. The hot air passed into the kiln by way of the accelerator hole and chimneys, thereby replacing the cooler air inside the kiln. The accelerator hole was closed when the smoke temperature was approximately 120 °C so that the chimneys could perform their normal function thereafter. The kiln temperature was raised slowly until it reached 180-200 °C. The smoke then became thicker and whiter indicating the beginning of carbonization in the front and top of the kiln. The hot combustion air from the firing port was then limited by reducing the area of the firing port opening to about 6 cm by 6 cm. The firing port size was thereafter monitored and adjusted to allow for proper cold air inlet by observing the emitted smoke until complete carbonization took place. The smoke turned bluish when the kiln temperature was greater than 450 °C and then became more clear at the end of the carbonization process, when the temperature was higher than 500 °C. Each chimney of the kiln was sealed when the tar inside the outlet of the chimney was dry and hardened. The run was completed when all chimneys were sealed. The kiln required about 2 days for cooling.

Mud Beehive Kiln (MBH)

• Construction: The MBH kiln used in these experiments was typical of the charcoal kilns used in rural areas of Thailand. The material used for construction of the MBH kiln wall was a soft lateritic mud or sandy clay. The kiln floor was dug below ground level. All of the wood was

placed in the pit. Small pieces of wood, grass, and leaves were placed on top of the bundle. This pile was then covered with 10-15 cm of mud. The kiln wall was compacted by pounding with a flat piece of wood. The soft mud wall dried and hardened after a few days and was then self-supporting. Cracks in the dried mud wall were sealed with a slurry of mud and water. The design also included a firing port, one accelerator hole, and two chimneys. See Figures A-3, A-4, and A-5.

• Operation: see Brick Beehive Kiln (BBH) Operation (above)

Single Drum Kiln (SD)

- Construction: The single drum kiln used in these experiments was made from a typical 200 liter oil drum. It came from the Philippines where it was primarily used to produce charcoal from coconut shells. Because of its small size and durability, it is a mobile kiln. The drum was 90 cm high with a 57 cm diameter. There were four air inlet holes in the base of the drum, each 2.5 cm in diameter. Air inlet holes of the same diameter were also placed in the side of the drum 5 cm, 35 cm, and 65 cm up from the base of the drum. See Figures A-6 and A-7.
- Operation: The operation of the SD kiln began with combustion and was followed by carbonization with direct draft. The fire was set at the bottom of the kiln. The wood was then loaded into the top of the drum. The kiln air inlet holes were plugged when the smoke emitted from the chimney became more clear, indicating that the transformation to charcoal had been completed. The cooling period was about 6 hours.

Rice Husk Mound Kiln (RHM)

- Construction: This kiln was made by simply digging a shallow pit in the ground. The wood was loaded horizontally into the pit and covered with rice husks. The RHM kiln did not require a chimney. See Figures A-8 and A-9.
- Operation: The fire was lit at the bottom of the piled wood. The entire wood pile was allowed to burn with free access to air until the wood bark was in flames. This required about 1 hour. The wood pile was then covered with about 6 inches of rice husks. The supply of air to the carbonization reaction was achieved via the porous nature of the rice husk pile. At the end of the carbonization process, the smoke became more colorless. The kiln was then sealed with a covering of soft, sandy soil or a metal sheet and sealed with clay. Two or three days were required for cooling.

Earth Mound Kiln (EM)

• Construction: This kiln was also made by simply digging a shallow pit in the ground. The wood was loaded horizontally into the pit and covered with grass or leaves and then earth. The layer of grass or leaves was thick enough to prevent the earth from directly touching the pile of wood. An opening was left in the earth mound covering for a chimney. See Figure A-8.

• Operation: The operation of the EM kiln was similar to that of the RHM kiln. The entire wood pile was covered with grass or leaves and then earth, except for one area required for lighting, located opposite of the chimney. The fire was lit at the opening with a small amount of firewood. The pile was then allowed to burn with free access to air until the wood bark was in flames. At this point the flaming end was covered with earth. Reverse draft and control of the air supply was achieved through the chimney. At the end of the carbonization process, the chimney was removed and the hole was covered with earth. The kiln was then sealed in the manner as the RHM kiln.

Age of Kilns

The beehive kilns used in these experiments were mature, having been fired by CRC many times previous to our use of them. They thus represent a more typical case than would newly made kilns, which take some time for their performance to stabilize. The single drum kiln was a newly made device, but of an old drum as is the common practice in the field. Rice husk and earth mound kilns, of course, are only used once since they are destroyed in the process of unloading them.

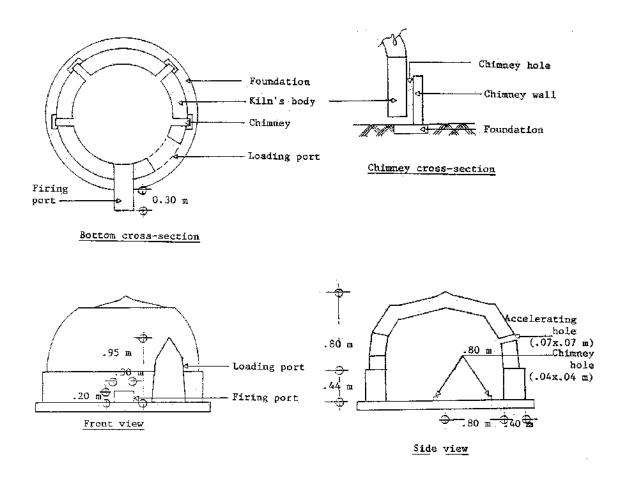


Figure A-1. Diagram of the brick beehive kiln (reproduced from Chomcharn, 1985).



Figure A-2. Brick beehive kiln at the Charcoal Research Centre.

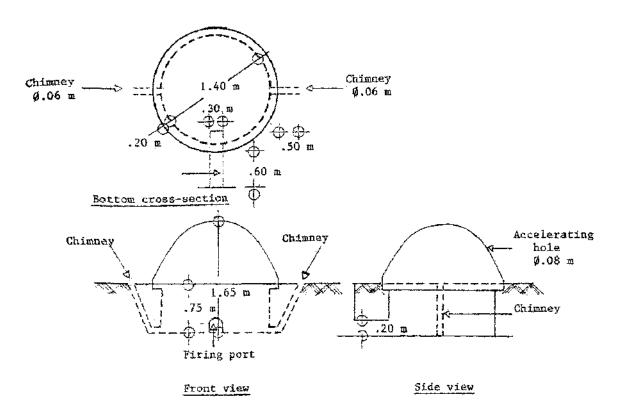


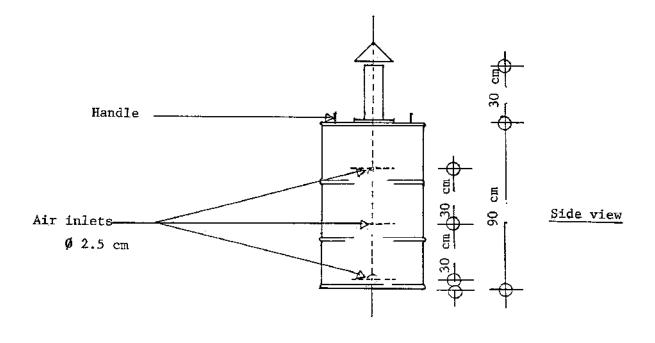
Figure A-3. Diagram of the mud beehive kiln (reproduced from Chomcharn, 1985).



Figure A-4. Ignition of the mud beehive kiln in the hood system at the Charcoal Research Centre.



Figure A-5. Mud beehive kiln inside the hood system at the Charcoal Research Centre.



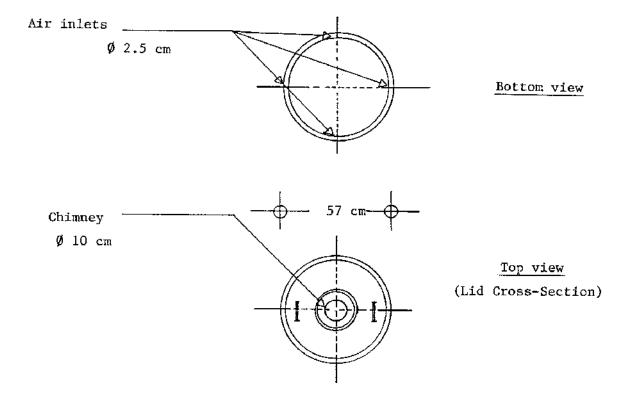


Figure A-6. Diagram of the single drum kiln (reproduced from Chomcharn, 1985).



Figure A-7. Single drum kiln at the Charcoal Research Centre.

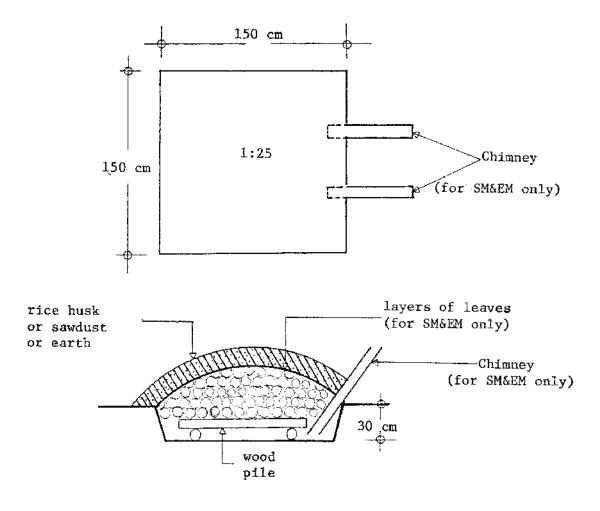


Figure A-8. Diagram of the earth mound and rice husk mound kilns (reproduced from Chomcharn, 1985).



Figure A-9. Ignition of the rice husk mound kiln at the Charcoal Research Centre (reproduced from Chomcharn, 1985).

Appendix B. Detailed Sampling Procedure

The sampling configuration, from upstream to downstream, consisted of a metal sampling probe, a TSP sampling cassette holding a 37-mm diameter quartz fiber filter (Whatman), a low-flow pump (SKC Aircheck Sampler, model 224-PCXR7), and an 80-liter Tedlar bag (SKC West, Fullerton, CA). These parts were connected with ¼" Teflon tubing. Figure B-1 shows a schematic of the sampling system used in the 15 kiln experiments.

80-liter Tedlar Bag Sampling: The duration and sampling flow rate of filling each large bag was recorded. An 80-liter bag (the mixing bag) was created by filling an empty 80-liter Tedlar bag with a time-weighted fraction of air from each filled sample bag. When only one 80-liter bag was required for a given run (when the firing time was less than 5 hours), that bag was treated as the mixing bag. The sampling parameters and mixing bag creation method for each kiln type are shown below:

- MBH and BBH: The firing time of these kilns was from 38 to 61 hours. A sampling flow rate of 100 ml/min was used, resulting in the filling of 4 or 5 large (80-liter) Tedlar bags per run. The duration of filling each bag was 12 hours. To create the integrated sample, one liter per two hours of filling was taken from each large bag and placed in the clean, empty 80-liter mixing bag (please see Table 1 for a sampling summary).
- EM: The firing time of the EM kiln was from 20 to 24 hours. At a sampling rate of 200 ml/min, 4 or 5 large bags were needed for each run. Each bag was filled for 5 hours. Two liters per hour of filling was taken for the mixing bag.
- SD and RHM: The firing time of the SD and RHM kilns was from 4 to 7 hours. Only one or two 80-liter bags were needed for each run.

<u>Use of small (1-liter) Tedlar bags</u>: These were used to capture samples from filled large Tedlar bags. Two small bags (replicates) were filled per mixing bag. These 1-liter bags were taken to the laboratory at KMUTT for GC analysis. For runs 1 and 3 for each kiln, 1-liter bag samples were only taken from the mixing bags (1 bag per kiln run). For run 2 of each kiln, samples were taken from the mixing bag and all of the 80-liter Tedlar bags that were filled throughout the run.

Replicate Sample: A parallel sample (replicate) was taken once for each kiln (during run 2). When conducting the parallel sampling, a "Y" shaped probe was used to allow for a pair of identical sampling trains running simultaneously. The purpose of the parallel sampling was to evaluate the method precision. See Table E-1.

Ambient Sample: A probe, filter cassette, pump, and 80-liter Tedlar bag was also used for ambient air sampling during one run of each kiln (run 1). This sampling was conducted at least head-height off the ground, upwind of the kiln area, and was completed before the firing of the kiln. A flow rate of 3 liter/min was used for 25 minutes to fill the large bag. With cassette alone, however, the sampling continued for another 2 hours to collect a large enough amount of TSP.

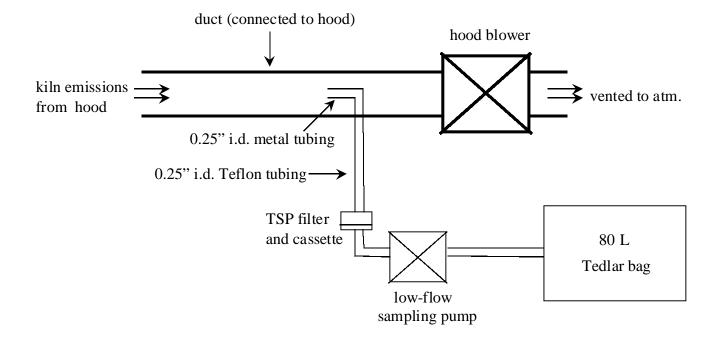


Figure B-1. A schematic of the sampling system used in the 15 kiln experiments.

<u>Canister Protocol</u>: From the mixing bag of each of the 15 runs, a pre-cleaned and evacuated 850 ml stainless-steel canister was filled to 2 atm using a battery-operated pump. In addition, during run number 2 for each kiln, one other canister was filled with an ascarite trap placed upstream of the canister. Use of the ascarite trap allows for more accurate N_2O analysis.

TSP Sampling Protocol: Whatman quartz fiber filters (37 mm diameter) were employed for collection of TSP from the charcoal kiln emissions. Before sampling, the filters were baked at 700°C in a furnace for at least 2 hours and then placed in a desiccator for at least 24 hours before weighing. The filters were weighed in a 5-place balance immediately after being taken out of the desiccator. Each weighed filter was placed in a labeled cassette, sealed with plastic caps, and transported to the site (CRC).

<u>1-liter Tedlar Bag Grab Samples</u>: A set of grab (one-time, non-integrated) hood samples were collected in 1-liter Tedlar bags for 9 of the 15 total kiln experiments. A ¼" Teflon tube attached to a wooden pole was used as the sampling probe. The inlet of the Teflon tube was placed about 0.5 m directly above the top of the kiln. This area above the kiln was well-mixed and full of smoke due to the large hood which contained and directed the emissions. (Without the hood, the smoke from all the kiln outlets would not be well-mixed and at relatively high concentration.)

The 1-liter grab samples were analyzed at KMUTT, as were the mixing bag, individual large bag, and ambient samples. The purpose of these grab samples was to investigate and develop a sampling method much simpler than the more complete (and cumbersome) integrated hood measurements. The similarity of the grab sample analytical results to those from the integrated measurements was evaluated (see results section). Such extended grab sampling may be useful for future field work (kiln sampling), obviating the difficult tasks of building and handling a large hood and duct system and making long duration measurements.

<u>Deposited Solids Emissions Measurements</u>: In order to quantify the amount of material from the emissions that deposited onto the inner wall surfaces of the hood and kiln, two patches made of galvanized zinc (3" x 3") were placed on each of these surfaces. Weight and surface area of each patch were measured before kiln firing. After kiln firing was completed, each patch was removed and weighed at KMUTT. The total mass of material deposited onto the hood surface and kiln walls was calculated based on the net weight increase of the patch and the ratio of the total hood or kiln surface area to the patch area.

Condensable Liquid (Condensables) Emissions Measurements: A metal U-tube was built into the emissions sampling ductwork system downstream of the large hood, prior to the gaseous sampling system to collect condensable species. This proved to be an unworkable method for estimation of the total amount of condensables ultimately emitted from the kilns. Therefore, two follow-up kiln experiments (one BBH and one EM) were performed in order to better quantify the condensables emitted. In these experiments, the sampling probe was placed directly over a kiln chimney and led to a series of two flasks immersed in water baths in order to more directly trap the condensable liquids.

A summary of the filter, bag, canister, and grab samples taken for each kiln run, along with firing times and sampling probe flow rate, is shown in Table 1.

Appendix C: Analytical Methods

<u>Solid Samples</u>: Calorific value and carbon (C) content were determined for two samples of each of the wood species (Eucalyptus camaldulensis and Leucaena leucocephala). Weight and moisture content were measured for each of the fifteen batches of wood used. Wood moisture content was determined by weighing several cross-sectional samples before and after oven drying (several hours at 110 °C).

After kiln firing was complete, unused firing wood, charcoal, brands, and ash were removed from the kiln and weighed. Samples of the charcoal, brands, and ash were removed and later analyzed for C content. Calorific value of the charcoal and wood was also determined. Carbon content of the TSP was analyzed using an automated thermal-optical method at Sunset Laboratory, Forest Grove, Oregon, USA (Johnson et al., 1981).

Gaseous Samples: Samples collected in 1-liter Tedlar bags were transported from the CRC to the laboratory at KMUTT for gas chromatography (GC) analysis of CO₂, CO, CH₄, and TNMHC. A Chrompack CP9001 GC along with a methanizer system interfaced to a Chrompack Flame Ionization Detector (FID) was employed for analysis of CO, CO₂, and CH₄. In this system, a Carbosphere (Alltech Co., USA) packed column was used to separate these three compounds. The separated CO and CO₂ were converted by the methanizer to CH₄ which was then detected by the FID. Total hydrocarbons (THC) were measured by the Chrompack GC/FID system equipped with a blank stainless-steel column (the air peak was subtracted). TNMHC was then determined indirectly by subtracting the CH₄ content from the THC peak. The GC was calibrated before each use via a calibration curve (at least 5 points) generated from locally made CO₂, CO, and CH₄ standards. The GC calibration was checked with a standard canister gas mixture of CO₂, CO, and CH₄ prepared by Scott Specialty Gases, Inc., USA

The filled stainless-steel canister samples were shipped to OGIST and analyzed for CO₂, CO, CH₄, TNMHC, and N₂O. OGIST used a GC/FID/methanizer method to analyze CO₂, CO, and CH₄. The canisters were analyzed for N₂O using a GC interfaced with an electron capture detector (Rasmussen and Khalil, 1980, 1981; Rasmussen et al., 1982).

Information about the stability of gases in the bags over time is presented in Table E-2. Analyses of duplicates (two small bags taken from one large bag) are presented in Table E-3.

<u>Filters</u>: After sampling, the filter cassette was re-capped and transported to the laboratory at KMUTT. Before weighing, the caps were removed, and the filter and cassette were placed in a desiccator for at least 24 hours. The filter was weighed immediately after removal from the desiccator. Weight, date, code, and balance number were recorded in a sampling sheet. The weighed TSP filter was then place in a labeled petri dish which was sealed in a plastic bag and stored in a refrigerator until being sent out for carbon analysis. The cassette was cleaned by wiping and could be reused. Table E-4 shows the weighings of laboratory and field blanks.

Appendix D: Follow-up Experiments to Determine Condensable Liquid Emissions

The results of these experiments are summarized in Table D-1.

Kilns Tested: One run each of BBH and EM kilns was performed using eucalyptus wood.

Parameters Measured:

• Airborne Measurements: CO₂, CO, CH₄, TNMHC

• Solid/liquid Measurements:

- Condensables (liquid): mass, carbon content

Wood: massCharcoal: mass

- Brands and ash: mass

Summary of Sampling Procedure:

- Dual sampling line system: Two sampling lines were utilized. The first line, called the condensables sampling line, consisted of 1/4 inch i.d. metal tubing, followed by two 2-liter Erlenmeyer flasks(capped with two-holed rubber stoppers), followed by a large protective quartz fiber filter, followed by a SKC Aircheck Sampler pump venting to the atmosphere. The sampling end of the metal tube was placed directly above one of the chimneys of the kiln. This sampling line pump ran continuously throughout the kiln experiments at 1.5 l/min. The large, protective filter was checked and replaced when visually full to prevent blockage of the line. The two Erlenmeyer flasks were immersed in water baths in order to keep the flasks cool to promote condensation. The bath water was changed periodically with fresh, cool stream water. The second part of the sampling line, called the gas sampling line, was connected via a "Y" connection to the condensables sampling line after the second Erlenmeyer flask and just before the large, protective filter. This sampling line consisted of 1/4 inch metal tubing, followed by a 37 mm Whatman quartz fiber sampling filter, followed by a SKC Aircheck Sampler pump, followed by a 80-liter Tedlar bag. The gas sampling line pump was programmed to collect emissions for 2 minutes out of every hour of kiln firing time. The gas sampling line pump flow rate was the same as that used in the condensables sampling line (1.5 1/min). The TSP sampling filter was checked periodically for particulate buildup to prevent line blockage. The TSP sampling filter was replaced if a large amount of particulate buildup was seen on the filter. Figure D-1 shows a schematic of the sampling system used in the follow-up experiments.
- Handling of the condensables (liquid): When the kiln firing was completed, the condensable liquids were weighed in the field (at the CRC). The condensables were transferred to a glass jar, sealed, and brought back to KMUTT for subsequent carbon content analysis as were the solid samples of the fuel, charcoal, and brands.

• Collection of kiln emission gases in Tedlar bags: At the end of the kiln run, three 5-liter metal-reinforced Tedlar bags (replicates) were filled from the large 80-liter Tedlar bags. These small bags were filled to a maximum of 3 liters to help minimize bag leakage. In addition, one ambient air sample was collected in a 5-liter metal-reinforced Tedlar bag for each of the two kiln experiments. The ambient samples were taken before kiln firing began, at a height of at least 1.5 meters above the ground, and away from human activity. These 5-liter bags were filled to a maximum of 3 liters. The bags were wrapped individually and placed in a hard case for shipment. GC analyses of the 5-liter bags were performed at EOHSI.

Table D-1. Data for follow-up experiments (determination of condensable liquid emissions)

	BBH	ЕМ
Total wet wood mass, kg	786	210
% wood moisture (dry basis)	20.2	19.4
Dry wood mass, kg	651	175
Charcoal mass, kg	230	53
Brands mass, kg	5.0	5.0
Ash mass, kg	1.5	4.0
C fraction of wood *	0.443	0.443
C fraction of charcoal *	0.789	0.748
C fraction of brands *	0.635	0.523
C fraction of ash *	0.089	0.048
Wood C, kg	288	77.6
Charcoal C, kg	182	39.6
Brands C, kg	3.2	2.6
Ash C, kg	0.10	0.20
C in gases and condensables, kg (by difference)	104	35.1
Charcoal C yield	0.63	0.51
Charcoal mass yield (dry basis)	0.35	0.30
Charcoar mass yield (dry busis)	0.33	0.50
Firing time, hr	39	19
Condensables sampling flow rate, L/min	1.5	1.5
Mass of condensables collected, g	13.8	2.59
Condensables emission concentration (g/m ³)	3.93	1.51
Carbon fraction of condensables	0.68	0.68
Condensables emission concentration (gC/m ³)	2.68	1.03
CO ₂ (ppm)	61300	16120
CO_2 (gC/m ³)	30.1	7.9
CO (ppm)	16280	3503
CH ₄ (ppm)	5122	696
TNMHC (ppm)	9633	954
CO/CO	0.266	0.017
CO/CO ₂	0.266	0.217
CH ₄ /CO ₂	0.0836	0.0432
TNMHC/CO ₂	0.157	0.0592
condensables/CO ₂	0.0889	0.130
K (sum of above 4 ratios)	0.595	0.449
CO ₂ (C) emitted (kg)	64.9	24.2
CO (C) emitted (kg)	17.2	5.27
CH ₄ (C) emitted (kg)	5.42	1.05
TNMHC (C) emitted (kg)	10.2	1.43
Condensables (C) emitted (kg)	5.77	3.14

(continued)

Table D-1. (continued)

	ВВН	EM
Percent of wood carbon in products:		
charcoal	62.94	51.10
brands	1.10	3.37
condensables	2.00	4.05
ash	0.0463	0.247
CO_2	22.51	31.25
CO	5.98	6.79
CH_4	1.88	1.35
TNMHC	3.54	1.85
Total	100.0	100.0

^{*} Average of values from the three original kiln experiments (carbon analyses not performed in these follow-up experiments)

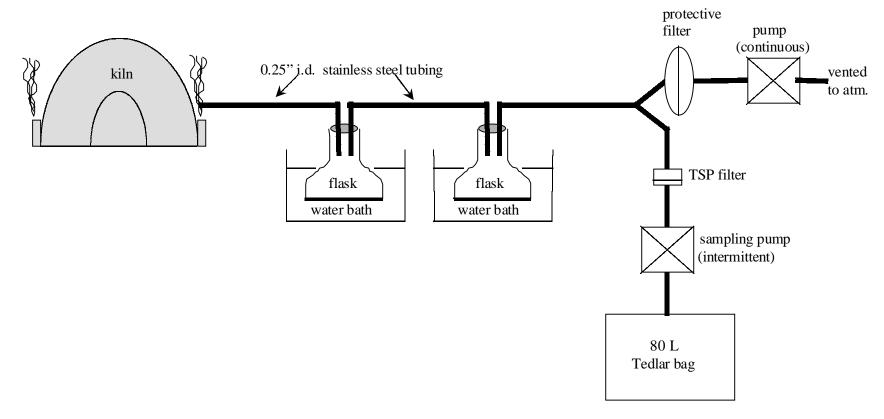


Figure D-1. A schematic of the sampling system used in the follow-up experiments.

Appendix E: Quality Assurance Plan

<u>Pilot Study</u>: Pilot work was conducted to: a) check the temperature at the sampling probe, b) check for leakage around lip of hood, c) check TSP build-up on filters, d) test the patch system and e) check for flow rate changes over the entire run period. Short-firing-time kilns (SD and RHM) were used in the trial runs.

For Each Kiln Run:

- Two sampling pumps were prepared before sampling by a) calibrating the pumps using a bubble flow meter, b) checking internal protection filters (they were changed if broken or discolored), and c) cleaning tubing, if discolored, with water and alcohol and drying.
- The Tedlar bags were cleaned by flushing with clean air at least three times before each use. The bags were checked to assure that there was no leakage.
- Several parameters were checked throughout the runs, including sampling flow rate, hood blower flow rate, and hood leakage. These parameters were checked near the beginning and end of each kiln run, as well as every 6 hours of firing for MBH and BBH, every 4 hours for EM, and every 2 hours for SD and RHM.

<u>Labeling</u>: Filters, large bags, small bags, etc. were carefully labeled. Sampling sheets were carefully filled out and maintained. The following labeling rules were used:

- The basic identifier on sampling sheets was "kiln type-run#", i.e., MBH-1, MBH-2, MBH-3, BBH-1, BBH-2, etc.
- Large Tedlar bags: LB-01, LB-02, etc. (the first large bag from run#1 for each kiln type was the ambient sample)
- Pumps: P-01, P-02, P-03
- Filters (code on cassettes and petri dishes): F-001, F-002, etc. (the first filter from run#1 for each kiln was the ambient sample)
- Small Tedlar bags:
 - MBH: MBH-1M (mixed bag from MBH run #1), MBH-2M (mixed bag from MBH run #2), MBH-2A (first integrated bag from MBH run #2), MBH-2B (second integrated bag from MBH run #2), MBH-2B-R (replicate of the second integrated bag sample from MBH run #2), MBH-2C, MBH-2D, MBH-2E (labels A through E represent each of the integrated 80-L Tedlar bag samples), MBH-3M (mixed bag from MBH run #3)
 - BBH: BBH-1M, BBH-2M..., the same as for MBH.
 - EM: EM-1M, EM-2M, EM-2A, EM-2B, EM-2B-R, EM-2C, EM-3M
 - SD: SD-1, SD-2, SD-2R, SD-3
 - RHM: RHM-1, RHM-2, RHM-2R, RHM-3
 - Ambient Samples: A-MBH-1, A-BBH-1, A-EM-1, A-SD-1, A-RHM-1.
 - Grab Samples: BBH-1-G1, BBH-1-G2, BBH-1-G3, BBH-2-G1, etc.
- Canisters: C-01, C-02, etc.; keyed to the kiln and run number in the sampling sheet

Method and Analytical Precision:

- One parallel, replicate sample (80-liter Tedlar bag and filter) was taken for one run of each kiln to assess the precision of the measurement method. These replicate analyses are shown in Table E-1. Three of the 1-liter mixed bag samples were run through the GC a second time after a one-week storage period in order to see the effect on the mixture of major gases. These data are shown in Table E-2. Also, two duplicate small Tedlar bags were filled from the large mixing bag for five of the kiln runs. This allowed for duplicate GC analyses at KMUTT. See Table E-3 for these data.
- In the local laboratory at KMUTT, two or more injections were made into the GC for each sample until a relative standard deviation (RSD) of less than 10% was obtained for all peak areas of interest. All GCs were calibrated before each use with at least five calibration points. The calibration curves for all measured compounds had a linear regression coefficient (R²) greater than 0.99. The concentrations of unknown samples fell into the calibration range for all species other than CO. In some instances, due to very high sample concentrations, the maximum CO calibration point was exceeded.
- After ten TSP filters were weighed, the first was reweighed. If the difference was more than 0.1%, all ten filters were reweighed. In addition, two laboratory blank filters and two field blank filters were used for each kiln run in order to check for any contamination during the transporting and handling processes and/or any changes in the analytical balance over time. The average field blank filter mass gained was subtracted from each of the sample filter masses. See Table E-4 for the blank filter weighings.

<u>Cross Laboratory Checks</u>: Results obtained from the local GC analyses (at KMUTT) were compared with results of canister samples analyzed by OGIST (see Results section). Although the KMUTT laboratory and the OGIST laboratory used their own locally made calibration standards on daily operation basis, each institution was provided with a calibration mixture of CO₂, CO, CH₄ from the same source (Scott Specialty Gases, Inc., USA). Thus, the locally made standards were calibrated with a common standard and calculations could be corrected based upon the common standard if there was any difference between a local standard and the common standard.

Back-Calculation of Volumetric Emission Rates:

The emission factor determination method used in this study was not designed to require knowledge or measurement of the volumetric emission rate of the kilns. The hood and blower system used in these experiments was designed to capture as much of the kiln emissions as possible, but can tolerate some leakage since the primary measures are the CO₂ ratios. It is important, however, that the flow rate is constant throughout. The flow rate of the single-speed hood blower, which was measured several times throughout each kiln run, was found to be quite constant. The range of average measured flow rates of the hood blower (for three runs of each kiln type) was 0.022-0.027 m³/sec.

Table E-1. Replicates. Two bags filled simultaneously through a y-probe.

	CO ₂ (ppm)	CO (ppm)	CH_4 (ppm)	TNMHC (ppm)
	X A	\ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \		11 1
BBH-2B-r1	21485	6897	2300	3307
BBH-2B-r2	21038	6839	2314	3153
MBH-2B-r1	4100	766	136	237
MBH-2B-r2	4115	755	131	228
SD-2M-r1	12801	5150	1410	2568
SD-2M-r2	11756	4141	1124	2899
EM-2A-r1	6747	2039	438	459
EM-2A-r2	6587	2010	431	458
RHM-2M-r1	13497	1056	224	377
RHM-2M-r1	13898	1106	248	394

Table E-2. Storage test data.

Sample/	CO_2	CO	CH_4	TNMHC	All Species	CO/CO_2	CH_4/CO_2	TNMHC/CO ₂	$(CO+CH_4+TNMHC)/$
date of analysis	(ppm)	(ppm)	(ppm)	(ppm)	(ppm)				CO_2
BBH-2M-b1									
5/13/96	16267	4818	1403	1365	23853	0.296	0.086	0.084	0.466
5/19/96	12157	4369	1277	1438	19241	0.359	0.105	0.118	0.583
MBH-1M-b1									
5/13/96	6541	1436	305	243	8525	0.220	0.047	0.037	0.303
5/19/96	4743	1281	274	285	6583	0.270	0.058	0.060	0.388
MBH-2M									
5/13/96	5411	981	223	249	6864	0.181	0.041	0.046	0.269
5/19/96	4172	911	216	232	5531	0.218	0.052	0.056	0.326

Table E-3. Analyses of duplicate small bags filled from the same large mixed bag

Name	CO_2	CO	CH_4	TNMHC
	(ppm)	(ppm)	(ppm)	(ppm)
BBH-1M-b1	23352	6730	2338	2382
BBH-1M-b2	26587	6602	2367	2187
BBH-2M-b1	16267	4518	1403	1365
BBH-2M-b2	15459	4594	1454	1357
BBH-3M-b1	26742	6241	2187	3027
BBH-3M-b2	28801	5962	2236	2934
MBH-1M-b1	6541	1436	305	243
MBH-1M-b2	6333	1407	295	289
MBH-3M-b1	6986	1151	329	423
MBH-3M-b2	7540	1143	327	403

Table E-4. Field and laboratory blank filter data. The average of the field blank values was subtracted from the real TSP samples. The laboratory blank data were not used for correction.

Field (CRC) Blanks	Filter #	Mass Change (mg)
BBH-1	F-047	0.010
BBH-2	F-012	0.045
BBH-2	F-013	0.055
BBH-3	F-052	0.175
BBH-3	F-053	-0.210
MBH-1	F-016	0.120
MBH-1	F-017	0.055
MBH-2	F-031	0.040
MBH-2	F-034	0.025
MBH-2	F-035	0.020
		mean = 0.034
Laboratory Blanks		
BBH-1	F-041	-0.015
BBH-2	F-001	0.065
BBH-2	F-002	0.060
BBH-3	F-054	-0.025
MBH-1	F-014	-0.020
MBH-1	F-015	0.150
MBH-2	F-032	-0.035
MBH-2	F-033	0.060
		mean = 0.030