SAFARI 2000 Gas Emissions from Biofuel Use and Production, September 2000

Abstract

Domestic biomass fuels (biofuels) were recently estimated to be the second largest source of carbon emissions from global biomass burning. Wood and charcoal provide approximately 90% and 10% of domestic energy in tropical Africa, respectively. As part of the Southern Africa Regional Science Initiative (SAFARI 2000), the University of Montana participated in both ground-based and airborne campaigns during the southern African dry season of 2000 to measure trace gas emissions from biofuel production and use and savanna fires, respectively.

In September 2000, open-path Fourier transform infrared spectroscopy (OP-FTIR) was used to quantify 18 of the most abundant trace gases emitted by wood and charcoal cooking fires and an earthen, charcoal-making kiln in Zambia. These are the first, in-situ measurements of an extensive suite of trace gases emitted by tropical biofuel burning.

This data set provides biofuel burning emission ratios and emission factors from field measurements for the following trace gases (in order of abundance): carbon dioxide (CO2); carbon monoxide (CO); methane (CH₄); acetic acid (CH₃COOH); methanol (CH₃OH); formaldehyde (HCHO); ethene (C₂H₄); ammonia (NH₃); acetylene (C₂H₂); nitric oxide (NO); ethane (C₂H₆); phenol (C₆H₅OH); propene (C₃H₆); formic acid (HCOOH); nitrogen dioxide (NO₂); hydroxyacetaldehyde (HOCH₂COH); and furan (C₄H₄O).

The investigators also measured trace gases from savanna fires with an airborne FTIR (AFTIR) on the University of Washington Convair-580 research aircraft in August/September of 2000. The data set for AFTIR measurements is archived at ORNL DAAC at <u>ftp://daac.ornl.gov/data/safari2k/</u> atmospheric/fire_emissions/.

Background Information

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Project: SAFARI 2000

Data Set Title: SAFARI 2000 Gas Emissions from Biofuel Use and Production, September 2000

Site: Kaoma, Zambia (and vicinity) Westernmost Longitude: 24.82° E Easternmost Longitude: 24.82° E Northernmost Latitude: 14.86° S Southernmost Latitude: 14.86° S

Data Set Citation:

Yokelson R. J. 2004. SAFARI 2000 Gas Emissions from Biofuel Use and Production, September 2000. Data set. Available on-line [http://daac.ornl.gov/] from Oak Ridge National Laboratory Distributed Active Archive Center, Oak Ridge, Tennessee, U.S.A.

Web Site: https://www.umt.edu/chemistry/faculty/yokelson.htm

Data File Information

The ground-based campaign measured trace gas emissions from the production and use of biofuels. The data files from the ground sampling efforts are listed below:

charc_fire_emiss_factors.csv charc_fire_emiss_ratios.csv charc_kiln_emiss_factors.csv charc_kiln_emiss_ratios.csv wood_fire_emiss_factors.csv
wood_fire_emiss_ratios.csv

All emission factors units are grams of compound emitted per kilogram of dry fuel. Emission ratios are unitless.

The files are ASCII text files in comma-separated-value format. The columns are individual fires from this study, or published values from previous studies. The number of fires from this study and previous studies vary for each biofuel type, but the rows, which are the measured constituents from each, are mostly of a common set. The definitions for these constituents can be found in the table of chemical formulas listed in the companion file. Note that the delta symbol in the parameter fields for emission ratios has been replaced by 'D's in the text files. Blank fields in the data files indicate the measurements were below the detection limit for the FTIR data. Note, however, that blank fields for other related studies reported in the data files only indicate that the constituent was not part of that particular study.

Trace Gas Chemical Name	Chemical Formula
carbon dioxide	CO ₂
carbon monoxide	СО
methane	CH ₄
acetylene	C ₂ H ₂
ethene	C ₂ H ₄
ethane	C ₂ H ₆
propene	C ₃ H ₆
furan	C ₄ H ₄ O
acetic acid	CH ₃ COOH
methanol	CH ₃ OH
phenol	C ₆ H ₅ OH
formaldehyde	НСНО

Chemical formulas and names used in the data files are provided below:

formic acid	НСООН
hydroxyacetaldehyde (other names include glycolaldehyde and hydroxyethanal)	HOCH ₂ CHO
ammonia	NH ₃
nitric oxide	NO
nitrogen dioxide	NO ₂
Other Terms used	Denoted as
non-methane organic compounds	NMOC
grams of carbon contained in all the NMOC detected	gC(NMOC)

The table below defines some of the terms commonly used in the discussion of biomass burning in general and in this documentation file specifically. (Also see <u>http://www.umt.edu/chemistry/faculty/yokelson/firetermbob.htm</u>.)

Biomass Burning Term	Definition
Biofuel	This term has been used most often to denote biomass that is used as a (renewable) domestic or industrial energy source (cooking, heating, lighting, etc.) instead of fossil fuels. It may confuse people to use "biofuel" as a term equivalent to biomass.
Emission Factor (EF)	Grams compound emitted per kg of fuel burned (on a dry mass basis). EF * fuel consumption * burned area = the total emissions of a compound from a burned area.
Emission Ratio (ER)	Dimensionless molar ratio between two emitted compounds measured at a fire. Most often reported as $\Delta X/\Delta Y$ where " ΔX " and " ΔY " refer to excess mixing ratios (defined as the mixing ratio of a species in the smoke minus the mixing ratio of that species in the background air) and "Y" is usually CO or CO ₂ .

Combustion Efficiency (CE)	Defined as the fraction of fuel carbon converted to carbon as CO_2 . Historically, combustion engineers want the CE of their reactors to equal 1 to maximize energy delivered by the fuel.
Modified Combustion Efficiency (MCE)	Defined as $\Delta CO_2/(\Delta CO_2 + \Delta CO)$ and abbreviated as MCE. Related to CE, but easier to measure in the real world. MCE has a further advantage, in that other carbonaceous compounds can be plotted vs. MCE as independent variables. Pure flaming usually has MCE of 0.97-0.99, and pure smoldering usually has MCE of 0.75-0.85. CE and MCE are higher when the ratio of flaming to smoldering combustion is higher.

Introduction

To help address crucial gaps in our knowledge of African fire emissions, the University of Montana participated in both ground-based and airborne dry season campaigns as part of SAFARI 2000. The ground-based campaign focused on the trace gas emissions from the production and use of biofuels (Bertschi et al., 2003a) and the unlofted emissions sometimes produced by prolonged smoldering combustion in wooded savannas (Bertschi et al., 2003b).

Measurements

Wood Cooking Fires

In rural areas of Zambia, open wood fires are the main source of heat for cooking. Emissions from three open wood cooking fires that were built and tended by local residents in both Kaoma (1 fire) and the remote village of Milumbwa (2 fires; location ~ 50 kilometers west of Kaoma on a one-lane dirt road) were measured. The cooking fire in Kaoma was in a sheltered area of the backyard of our rented house and the Milumbwa cooking fires were conducted inside traditional huts (~ $2 \times 2 m$ floor area). The fires in both locations were built with the regions most commonly used firewood (the miombo tree species

Julbernardia sp. and *Brachystegia* sp.). The fuel was typically 3-5 pieces of small diameter (2.5-7.0 cm) wood branches or logs with their long axes arranged radially. The fires were ignited at the center of the group where they burned gently (to minimize wasted heat or wood). The wood was pushed towards the center as the fire burned and quickly extinguished after cooking. At both locations, the OP-FTIR was supported by shipping crates about 1 m above the fires and weak convection from the fires directed the emissions through the optical path of the open White cell. Spectra were acquired continuously over the course of each fire with each individual spectrum lasting 3-11 seconds. Background spectra were collected in clean air upwind of the fires before and after each fire. A hand-held, chromel-alumel thermocouple and a portable, digital barometer (Cole-Parmer) were used to measure the temperature and pressure, respectively, in the OP-FTIR optical path. AC power was available for the FTIR and computer in Kaoma, but were powered by automotive batteries in Milumbwa and other rural locations.

Charcoal Cooking Fires

Charcoal is used mostly in urban areas and it is normally burned in a rudimentary stove or metal container. Emissions were measured from one charcoal cooking fire in Kaoma. The charcoal was produced locally from the same tree species used for firewood above and it was burned in a simple stove consisting of an open, perforated metal cylinder supported by three legs and underlain by a 'floor' to catch ashes. The OP-FTIR deployment was the same as for the wood cooking fires (described above).

Earthen Charcoal-Making Kiln Fires

Charcoal is produced in rural areas for use mainly in urban areas. Our kiln study site was in a remote clearing in the miombo woodland about 20 km southeast of Kaoma (14.86 S, 24.82 E). Local residents experienced in charcoal making constructed the kiln and tended it throughout the experiment. The wood used for making the charcoal (*Julbernardia* sp. and *Brachystegia* sp.) was felled nearby and cut into logs 1-2 m in length. After drying, the logs were stacked about 1.5 m high. Soil clumps (approximately 40 x 40 x 20 cm) held together by dry grass roots were excavated next to the wood stack and used to encase the wood in an earthen layer 20 to 40 cm thick. This small kiln

was designed to produce 5 bags of charcoal weighing about 40 kg each. The wood was ignited through a small opening at one end of the kiln on the morning of the 10th of September. A few minutes after ignition, the opening was sealed with more soil clumps to reduce airflow into the kiln and promote carbonization of the wood charge. On the morning of September 14th, the kiln was broken to retrieve the charcoal. The kiln emissions emanated from numerous small holes in the walls and they were conveniently directed horizontally by a strong easterly wind through the optical path of the OP-FTIR system, which was operated as described for wood cooking fires. Background spectra were collected in a smoke-free area upwind of the kiln before and after each smoke measurement. Because of the need to sample both cooking fires and kiln emissions within one week and at different locations, it was not possible to measure the kiln emissions continuously. Instead, 3 extended measurements of the emissions were collected, lasting about 1-2 hours each, on the 10th, 11th, and 13th of September, which spanned the 4-day lifetime of the spectra from this study.

The photograph below, oriented upwind, shows the smoke drifting sideways out of the kiln and through the optical path of our open-path FTIR trace gas measurement system.



Additional photographs that complement the ground-based OP-FTIR measurements are available at <u>http://daacsti.ornl.gov/S2K/s2k_photo_pages/main_photos.html</u>.

Calculation of Emission Ratios and Emission Factors

The excess mixing ratios (ΔX , the mixing ratio of species 'X' in the smoke minus the mixing ratio of 'X' in the background air) that are observed in biomass-burning studies reflect the degree of dilution of the smoke at the point of measurement. Thus, a more widely-used, derived quantity is the normalized excess mixing ratio where ΔX is compared to a simultaneously-measured smoke tracer such as ΔCO or ΔCO_2 . A measurement of $\Delta X/\Delta CO$ or $\Delta X/\Delta CO_2$ made in nascent smoke (< ~5 minutes old) is an emission ratio (ER). Fireintegrated excess mixing ratios were used to calculate fire-average emission ratios

This data set contains data files of fire-average ER for all wood cooking, charcoal cooking, and kiln fires, as well as the study-average ER. For the

charcoal kiln data, an ER for each day was calculated and the daily measurements were averaged to obtain the kiln-average ER. Also included in the data files are the previously measured ER for biofuel use to facilitate comparisons with the results of this study.

Four previous studies measured some of the emissions from open wood cooking fires (Brocard et al., 1996; Smith et al., 2000; Kituyi et al., 2001; and Ludwig et al., 2002). Kituyi et al. (2001) measured trace gas emissions from wood burned in both open fires and different types of stoves. They found that the use of various stoves strongly influenced the mix of trace gases emitted. At least four previous studies quantified some trace gas emissions for charcoal cooking fires (Kaoma and Kasali, 1994; Brocard et al., 1996; Smith et al., 2000; Kituyi et al., 2001), and emission ratios for earthen charcoal-making kilns were reported in three previously published studies (Lacaux et al., 1994; Smith et al., 2000; Pennise et al., 2001).

Emission factors (EF, g compound emitted per kg dm fuel burned) were calculated for each trace gas emitted from the wood cooking fires using the carbon mass balance method, which is described in full in Ward and Radke (1993). The carbon mass balance method cannot be used to calculate kiln EF in the same way that was done to calculate cooking fire EF because the carbon content changes during the kiln lifetime and much of the burned carbon is not volatilized. Justification for the method used can be found in Bertschi et al. (2003).

Results

The investigators estimate that Zambian savanna fires produce more annual CO_2 , formic acid, and NO_x than Zambian biofuel use by factors of 2.5, 1.7, and 5, respectively. However, biofuels contribute larger annual emissions of methane, methanol, acetylene, acetic acid, formaldehyde, and ammonia by factors of 5.1, 3.9, 2.7, 2.4, 2.2, and 2.0, respectively. Annual CO and C_2H_4 emissions are approximately equal from both sources. Coupling these data with recent estimates of global biofuel consumption implies that global biomass burning emissions for several compounds are significantly larger than previously reported. Biofuel emissions are produced year-round, they disperse

differently than savanna fire emissions, and they could strongly impact the tropical troposphere.

Compared to previous work, emissions of organic acids and ammonia in this data set are 3-6.5 times larger. Another significant finding is that reactive oxygenated organic compounds account for 70-80% of the total nonmethane organic compounds. For most compounds, the combined emissions from charcoal production and charcoal burning are larger than the emissions from wood fires by factors of 3-10 per unit mass of fuel burned and ~2 per unit energy released.

Additional Sources of Information

Related Data Sets

The investigators also measured stable and reactive trace gases from savanna fires with an airborne FTIR (AFTIR) on the University of Washington Convair-580 research aircraft in August/September of 2000 during the SAFARI 2000 dry season aircraft campaign in Southern Africa. The data set for AFTIR measurements is archived at ORNL DAAC at <u>ftp://daac.ornl.gov/data/safari2k/atmospheric/fire_emissions/</u>.

A detailed description of the airborne FTIR (AFTIR) sytem and the groundbased Open Path FTIR (OP-FTIR) system can be found in Yokelson et al. (2003) and Bertschi (2003a), respectively.

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