

Fine Particle and Gaseous Emission Rates from Residential Wood Combustion

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Residential wood combustion emissions were analyzed to determine emission rates and to develop chemical emissions profiles that represent the appliances and woods typically used in wood-burning communities. Over 350 elements, inorganic compounds, and organic compounds were quantified. A range of 4–9 g/kg dry fuel of particulate matter (<2.5 μm) and 5–22 g/kg volatile organic compounds were observed. Samples were collected using a dilution stack sampler equipped with a 2.5-μm particle selective cyclone. Emissions were diluted 20–70 times, cooled to ambient temperature, and allowed 80 s for condensation prior to collection. Wood type, wood moisture, burn rate, and fuel load were varied for different experiments. Fine particle and semivolatile organic compounds were collected on filter/PUF/XAD/PUF cartridges. Inorganic samples and mass were collected on Teflon and quartz filters. Volatile organic carbon compounds were trapped with Tenax (C₈–C₂₀), canister (C₂–C₁₂), and 2,4-dinitrophenylhydrazine impregnated cartridges (carbonyl compounds). Analysis of particle and semivolatile organic species was conducted by gas chromatography/mass spectrometry. Teflon filters were analyzed for mass by gravimetry, trace elements were analyzed by X-ray fluorescence, and ammonium was analyzed by automated colorimetry. Quartz filters were analyzed for organic and elemental carbon by thermal/optical reflectance, and ions were analyzed by ion chromatography. Select quartz filters were analyzed by accelerator mass spectrometry for carbon-12 and carbon-14 abundance. Canister and Tenax samples were analyzed by gas chromatography with a flame ionization detector, and carbonyl compounds were analyzed by high-performance liquid chromatography.

Introduction

Residential wood combustion (RWC) contributes to the ambient concentration of fine (PM_{2.5}) particulate matter (1–3) and volatile organic compounds (VOCs) (4), especially in residential neighborhoods during winter. Using 1997 emission inventory data, annual emissions from wood stoves in the United States are estimated to be composed of 368 000 tons of PM₁₀ and 527 000 tons of VOCs (5). In residential areas where wood is the predominant heating fuel, wood stoves have been shown to contribute as much as 80% of the ambient fine particle concentrations during winter (6).

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Previous studies on the organic composition of wood combustion products have focused predominately on the emissions of polycyclic aromatic hydrocarbons (PAHs) due to public health concerns. Some PAHs, such as benzo[*a*]pyrene, are known carcinogens (7), and Watts et al. (8) showed that ambient air mutagenicity and the concentration of PAHs were positively correlated with wood smoke-impacted areas. Wood combustion has been estimated to account for more emissions of PAHs than any other source (9).

Other wood combustion emissions studies have focused on the identification of compounds that distinguish wood-burning contributions from those of other sources. Elements, ions, nitrate, sulfate, ammonia, and total organic (OC) and elemental carbon (EC) are the most commonly used chemical species in past source apportionment studies. Within these species, contributions from vegetative burning, meat cooking, and other sources are based upon the relative abundance of water-soluble potassium, chloride, sulfate, OC, and EC in source and ambient samples. Similarities between meat-cooking and wood-burning constituents, as well as a commonality of EC+OC with those of motor vehicle exhaust, have resulted in large uncertainties in source contribution estimates (10, 11). Recent receptor modeling studies (3, 11) that have included measurement of organic compounds in addition to the more commonly measured chemical species have improved estimation of source contributions with respect to carbonaceous particles.

In this study, we measured more than 350 chemical species, including volatile and semivolatile organics from C₂ to C₂₀, particle-phase organics such as PAHs, methoxylated phenols, organic and elemental carbon, inorganic species, elements, and carbon-14. Fuels, appliances, and burning conditions were typical of residential wood combustion communities such as Denver, CO, and Reno, NV. Fuels and appliances included softwoods (gymnosperms), hardwoods (angiosperms), and synthetic logs in a fireplace as well as hardwoods burned in a wood stove. The objective of our study was to quantify emission rates of compounds, or classes of compounds, that may further improve assessment of source apportionment relative to estimates developed from previous studies. The ultimate goal of our study was also to increase the knowledge base of air toxics that are emitted from the combustion of wood during residential heating. The source profiles derived from these data were applied to the ambient fine particle source apportionment during the 1997 Northern Front Range Air Quality Study (NFRAQS) (11). These source measurements are compared to the emission rates developed from previous studies.

Experimental Methods

Sampling. Wood combustion emissions have been shown in previous studies to be highly variable and dependent on many factors related to burn conditions, fuels, and appliances (6, 9, 12–14). Some of the most important factors that have been shown in previous studies to affect these emission rates and compositions are type of appliance [wood stove (catalytic or conventional) versus fireplace], burn rate (related to amount of wood being burned and stove ventilation), type of wood [hardwood (angiosperm) versus softwood (gymnosperm)], configuration (grate versus no grate), and moisture content (high moisture if greater than 20% wet weight). An effort was made during the current study to address many of these parameters by including them in the tests conducted shown in Table 1. There were 19 individual wood combustion tests made (Table 1), including three replicates of pine burned in a fireplace, three replicates of dry oak burned in a wood

TABLE 1. Wood Combustion Test Conditions and Emission Rates for Carbon Monoxide, Total Volatile Organic Carbon (VOC), and Fine Particulate (<2.5 mm) Mass (PM)

sample	used in profile ^a		appliance	fuel	type of wood	grate	target		burn rate (kg/h) ^c	fuel burned (kg)	av firebox T (°C)	fuel moist. (% wet)	CO (g/kg) ^d	VOC (g/kg) ^d	PM (g/kg) ^d
	SVOC/PM ^b	VOC					fuel load	burn rate							
FS01	yes	yes	fireplace	Ponderosa pine	softwood	yes	na ^f	na	2.6	10.9	352.4	10.0	e	5.6	5.8
FS02	yes	no	fireplace	Ponderosa pine	softwood	yes	na	na	3.2	14.1	338.9	8.9	e	e	4.9
FS03	yes	yes	fireplace	Ponderosa pine	softwood	yes	na	na	2.8	13.1	298.6	9.5	e	5.8	4.7
FS04	no	yes	fireplace	pinion pine	softwood	no	na	na	2.7	12.5	346.7	8.1	e	5.1	9.0
FS05	no	yes	fireplace	pinion pine	softwood	yes	na	na	3.2	14.7	346.7	11.4	e	7.5	2.9
FH01	yes	yes	fireplace	mixed hardwoods	hardwood	no	na	na	3.8	14.3	266.9	13.1	e	8.1	4.2
FH02	yes	yes	fireplace	mixed hardwoods	hardwood	yes	na	na	3.0	10.7	404.2	13.3	e	7.9	5.5
FH03	yes	yes	fireplace	oak	hardwood	yes	na	na	2.2	10.0	335.0	14.9	e	14.5	6.6
FH04	no	yes	fireplace	mixed hardwoods	hardwood	yes	na	na	2.2	9.2	333.7	18.1	e	11.8	8.0
FH05	yes	yes	fireplace	oak	hardwood	yes	na	na	2.5	12.5	316.9	18.2	e	7.0	6.4
FSY1	yes	yes	fireplace	synthetic log	synthetic	yes	na	na	1.3	2.8	223.8	na	e	1.4	8.3
WH01	yes	no	wood stove	mixed hardwoods	hardwood	na	high	low	3.4	21.7	405.6	9.0	135.8	e	6.1
WH02	no	no	wood stove	mixed hardwoods	hardwood	na	low	high	6.2	11.7	519.3	14.1	178.6	e	2.5
WH03	yes	yes	wood stove	mixed hardwoods	hardwood	na	low	low	4.4	12.8	423.5	7.6	102.7	6.2	2.3
WH04	yes	no	wood stove	oak	hardwood	na	high	low	3.9	20.7	398.0	6.5	124.9	14.2	3.6
WH05	yes	yes	wood stove	oak	hardwood	na	high	low	4.1	21.9	402.9	6.5	104.8	15.2	4.4
WH06	yes	yes	wood stove	oak	hardwood	na	high	low	3.7	21.5	396.6	8.5	131.9	13.6	e
WH07	yes	yes	wood stove	oak	hardwood	na	high	low	5.6	22.5	474.7	15.2	131.9	55.3	7.2
WH08	no	no	wood stove	oak	hardwood	na	high	low	3.3	21.5	e	15.5	120.9	36.8	e

^a All samples collected are shown in this table. Not all of these samples were used to create the composite emission rate profiles shown in Table 2. ^b SVOC/PM, semivolatile and particulate chemical species measured. ^c Burn rate for wood combustion is in dry kg/h. ^d Emission rate calculated as g of pollutant/dry kg of fuel. ^e No data available. ^f na, not applicable.

stove, and two replicates of high moisture oak burned in the wood stove. The “high” moisture samples used are actually lower moisture levels than recommended by the EPA *Federal Register* (15) for conducting high moisture tests (20% wet weight). These woods were desiccated by the dry climate in Reno.

Wood combustion tests were conducted at Desert Research Institute (DRI) in Reno, NV, using wood samples obtained from Denver, CO. Fireplace tests were conducted in a Heatilator model E36 fireplace (with a model GR4 grate). Wood stove tests were conducted with a noncatalytic Pineridge appliance. Each appliance was placed in the center of a shed with a 18 ft by 8 in. i.d. stainless steel exhaust flue extended through the center of the roof. Wood types burned in the fireplace included Ponderosa pine, pinion pine, Missouri oak, scrub oak, mixed hardwood (cottonwood, birch, aspen), and synthetic log. Fires were started with black print newspaper. Kindling was the same species used as the fuel for every test. The weight of the fuel, and therefore the burn rate, was monitored continuously throughout all of the tests using a Digimatex scale. Wood moisture content was measured with a Delmhorst meter.

A three test-fuel charge was used for the fireplace tests, with two additions after the initial test charge. Addition of fuel occurred when one-third of the weight of the preceding test charge was reached. The fuel-loading density was maintained at 7.0 lb of wood/cf. firebox area (112.1 kg/m³) for all fireplace tests. Wood stove tests were conducted by igniting the fire, adding a 5–7 lb warm-up load, loading the desired fuel load [“high” and “low”, i.e., 10 and 3 lbs of wood/cf. firebox area (160.2 and 48.1 kg/m³)], and changing the air intake with the adjustable aperture on the face of the stove (high air, open 8 mm; low air, open 1 mm). The synthetic log test was conducted by placing one Duraflame log on the grate in the fireplace, lighting it, and letting it burn. Wood tests were terminated when 90% of the final test charge weight had been burned. Using this termination point, the smoldering condition characterized by a lower temperature burn with a lack of flame makes up approximately 20% of the fireplace burn cycle and approximately 35–40% of the wood stove tests.

The stack velocity for the fireplace tests was measured with an S-type pitot tube connected to a DP41-E Omega

pressure transducer. Stack gas and humidity were not measured, so default values of 19% oxygen, 1.8% carbon dioxide, and 2% moisture by volume were used to convert the differential pressure readouts from the pitot into velocities. These default values were obtained by Omni Environmental Services from fireplace tests using the same model Heatilator appliance that we used (16). Stack velocities measured were comparable to numbers obtained by previous tests conducted by Omni Environmental using the Heatilator appliance (16). Stack velocities for the wood stove tests were considerably less than those of the fireplace tests, and they were below the measurable range of the pitot tube. Wood stove stack velocity was calculated based on a model developed for the U.S. EPA (15). The model is based on a carbon stoichiometric relationship between carbon monoxide, carbon dioxide, and the fuel consumption. Flue gas samples were collected using an EPA Method 5H sampling train. Details of this system are explained elsewhere (15). Percent oxygen was measured electrochemically with a Teledyne model 920 gas analyzer. Percent carbon dioxide and carbon monoxide were both measured by infrared spectrometry with an NDIR model 702D gas analyzer. A three-point calibration with authentic gas standards was conducted before stove tests. Pre- and post-span checks were conducted on the analyzers to ensure analysis accuracy. The details about these model calculations, and the assumptions therein, can be found in the *Federal Register* (15).

A dilution stack sampler was built at DRI in order to collect emissions during the wood combustion source tests. The dilution sampler is a slightly modified version of the model built and described by Hildemann et al. (17). Modifications include improvements in portability and a longer mixing chamber (17 effective diameters as compared to 10) that provides more mixing time to ensure uniform sample distribution. Emissions were withdrawn from the fireplace or wood stove exhaust 8 ft above the bottom of the appliance through a 10 ft by 0.5 in. heated stainless steel line. Sample line flows ranged between 20 and 30 L/min, and flow was measured with a calibrated venturi meter purchased from Lambda Square, Inc. (Bay Shore, NY). Emissions were drawn into the dilution sampler, where they were mixed under turbulent conditions with ambient air that was filtered through a high-efficiency particulate air (HEPA) filter (to

remove particulate matter) and an activated carbon bed (to remove gas-phase organics) prior to entering the chamber. Fireplace emissions were diluted 20–40 times, and wood stove emissions were diluted 50–70 times. To assess the contribution of background chemicals to our wood combustion samples, system blanks were collected from dilution air flowing through the system at 1200 L/min with the sample inlet capped off. The background concentrations for all species ranged from approximately 50 to 500 times less than the wood combustion samples. Most of the analytes in the blanks were at or below the detection limit and thus were not subtracted from the wood combustion samples.

Samples were drawn through Bendix 240 cyclones located inside the dilution sampler at 113 L/min to remove particles larger than 2.5 μm and then collected on (a) Teflon-impregnated glass fiber filters (TIGF) followed by PUF/XAD/PUF cartridges for semivolatile/particle organics; (b) Teflon membrane filters for mass and elemental analysis; (c) quartz fiber filters for ions, carbon, and C-14 analysis; (d) stainless steel canisters for C₂–C₁₂ volatile organic compound (VOC) analysis; (e) Tenax-TA cartridges for C₈–C₂₀ hydrocarbon analysis; and (f) C₁₈ Sep-Pak (Waters, Inc.) cartridges impregnated with 2,4-dinitrophenylhydrazine (DNPH) for the analysis of carbonyl compounds. These collection methods are described in detail elsewhere by Zielinska et al. (18, 19).

Analysis Methods. Laboratory methods are described in detail elsewhere (18–20). Canister and Tenax-TA samples were analyzed for compound identification by high-resolution gas chromatographic separation with Fourier transform infrared/mass spectrometric detection (Hewlett-Packard GC/IRD/MSD, 5890/5965B/5970). High-resolution gas chromatography with flame ionization detection (GC/FID, Hewlett-Packard 5890 series II) was used for quantitative determination.

Oxygenated hydrocarbons measured with the canister and Tenax methods were quantified by GC/FID using relative response factors (also called effective carbon numbers, ECNs). The ECNs were developed for methanol, ethanol, ethyl acetate, furan, 2-furaldehyde, hexenal, and guaiacol using the method of Scanlon and Willis (21). Compounds that were not experimentally determined were calculated from their molecular structures using the method developed by Jorgensen et al. (22). The ECN correction factor will provide a lower limit for the levels of oxygenated compounds quantified as a result of losses of these compounds that occur during analysis. Potential areas for loss of oxygenated compounds during analysis include poorer penetration through the injection system or column when compared to nonpolar compounds. Loss may also occur from oxygenated compounds “sticking” to the walls of the canister during sampling and analysis (23). The degree of oxygenated compound loss from sampling and analysis was not determined during this study.

Carbonyl compounds collected on DNPH cartridges (as hydrazones) were eluted with 2 mL of HPLC grade acetonitrile and analyzed by HPLC (Waters, Inc.) with UV detection at 360 nm (18).

Prior to extraction of filter/PUF/XAD/PUF cartridges, deuterated internal standards were added to each filter–sorber pair for PAH compounds ranging from naphthalene-*d*₈ to coronene-*d*₁₂. PUF plugs were Soxhlet extracted separately with 10% diethyl ether in hexane, and the filter–XAD pairs were microwave extracted with dichloromethane. The samples were analyzed by the EI (electron impact) GC/MS technique, using a Hewlett-Packard 5890 GC interfaced to a 5970B mass selective detector (MSD) operated in selective ion monitoring (SIM) mode. Quantification of compounds extracted from the filter/PUF/XAD/PUF cartridge was conducted by comparing the response of the analyte to the

response of the deuterated internal standard that most closely matches its retention time and stability. To correct for the varying response factors encountered for compounds with different molecular structures, calibrations were created using authentic standards purchased from Aldrich Chemical Co. (Milwaukee, WI) and the National Institute of Standards and Technology (Gaithersburg, MD). Standards were injected for all of the PAH reported here except 1,7-dimethylnaphthalene, and the methoxylated phenols except ethylguaiacol and ethylsyringol. The compounds that were analyzed without authentic standards were quantified using the response factors of standards with similar structures.

Teflon filters were weighed prior to and after sampling to determine mass concentrations. These were analyzed for trace elements by X-ray fluorescence (XRF). A 0.56-cm² punch from the pre-fired quartz fiber filters was analyzed for organic and elemental carbon by the thermal/optical reflectance (TOR) method described by Chow et al. (24). Half of the quartz filter was extracted in 10 mL of deionized water, and the extract was analyzed for nitrate, sulfate, and chloride ions by ion chromatography; for ammonia by indophenol automated colorimetry; and for water-soluble potassium by atomic absorption spectrometry. Some of the quartz filters were analyzed at the University of Arizona–NSF AMS Facility by accelerator mass spectrometry for carbon-12 and carbon-14 isotopic abundance. These methods are described elsewhere (25). One-half of a quartz filter was submitted for ¹⁴C/¹²C analysis of wood combustion experiments taken in the fireplace for a hardwood, softwood, and synthetic log sample.

Results and Discussion

Particle and gaseous emission rates from wood burning are heavily dependent on the chemical composition of the fuels and the combustion conditions. The chemical composition of wood and the combustion process have been described previously (2, 4, 6, 9, 12, 26–34). The data presented here show results of wood combustion tests conducted in both a fireplace and a conventional (noncatalytic) wood stove.

Creation of Composite Wood Combustion Profiles. Since the primary objective of this study was to develop source profiles that represent major compositional differences in wood combustion, data are presented here in composite profiles corresponding to the major woods and appliances tested. These composite profiles were created from the emission profiles of the individual experiments shown in Table 1 with the aid of a cluster analysis. Clustering is a multivariate statistical technique of grouping data together that share similar values. A hierarchical cluster using Ward's Method from the statistical analysis software JMP (35) was utilized to exploit differences and similarities in the composition of individual wood combustion profiles (normalized by weight fraction). Hierarchical cluster data are often presented in a dendrogram in which similar data are branched together in closer proximity than less similar data. The cluster dendrogram shown in Figure 1 shows similarities/differences between the emission profiles of softwoods (FS01–FS04), hardwoods (FH01–FH03, FH05), and synthetic logs (FSY1) burned in a fireplace as well as hardwoods burned in a wood stove (WH01–WH05, WH07). The individual profiles used in the cluster analysis are the same profiles that are shown in Table 1 to be included in the profile development. The major differences in profile composition observed in this study can be attributed to the appliance used (wood stove versus fireplace) and the type of wood (softwoods, hardwoods, synthetic log). The compositional differences observed that led to the pooling of these data into composite profiles are discussed throughout the Results and Discussion section.

The high moisture fireplace sample (FH05) was separated from the other fireplace/hardwood samples in Figure 1. This

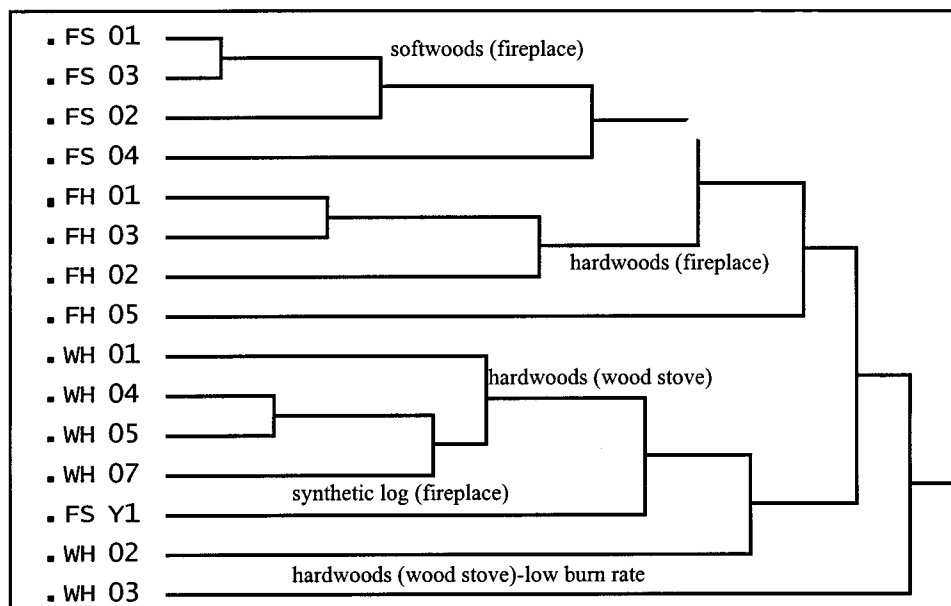


FIGURE 1. Hierarchical clustering of wood combustion source samples including fireplace tests for hardwoods, softwoods, and a synthetic log as well as wood stove tests with hardwoods.

sample was distinguished by its higher proportion of semi-volatile organics as compared to tests FH01–FH03 and was included in the fireplace/hardwood composite. The wood stove tests with low burn rates (WH02, WH03) were separated from the cluster of wood stove tests with higher burn rates. The low burn rate samples were distinguished by their higher amounts of semivolatile organic compounds. While WH03 was included in the wood stove composite profile, WH02 was not included because of an incomplete data set. Within the four composite profiles developed, the relative chemical composition is similar. Emission rates, however, were effected by both moisture content and burn rate. Differences in the emission rates for total VOC and fine particle mass are shown in Table 1 and discussed in a further section.

Table 2 shows average emission rates in mg of pollutant/kg of dry fuel for both softwoods and hardwoods burned in the fireplace, a synthetic log burned in the fireplace, and hardwoods burned in the wood stove. These rates are composite averages of the tests indicated in Table 1 and Figure 1. As indicated in Table 1, not all of the tests conducted are included in the composite profiles. Only tests where complete data sets exist were included in the composites. Measurement uncertainties used for composite source profiles are the larger of the 1σ standard deviations between tests or the root mean square of the analytical measurement uncertainties [SQRT ((replicate precision \times analyte concentration)² + (analyte detection limit)²)]. Differences between the individual tests are reflected in the profile uncertainties, and the individual emission rates for each test are available upon request.

Chemical Composition of Wood Smoke. Table 2 shows an abbreviated list (full list available in the Supporting Information or from the correspondence author) of the emission rates of individual fine particulate and gaseous species quantified during this study. A comparison of the data to previous studies is also provided. A summary of the emission rates for each respective chemical class is also shown in Table 2. These major chemical classes are discussed below.

Fine Particle and Gaseous Emission Rates. Total fine particle PM_{2.5} emissions rates for all wood combustion tests are summarized in Table 1. The composite emission rates were 5.1 g/kg for softwoods burned in the fireplace, 5.7 g/kg for hardwoods in the fireplace, and 4.7 g/kg for the wood stove emissions. The synthetic log yielded emissions of 8.3

g/kg. Varying the appliance, fuel, and burn conditions did not seem to have a large effect on the amount of fine particle emissions during this study.

The VOC emissions are defined here as any hydrocarbon, including oxygenated, which are measured by the canister, Tenax, or DNPH methods. For reporting the total VOC (Table 1, Table 2, Figure 2), the sum of all compounds measured by the canister analysis was used to enable comparison to previous VOC studies on wood combustion. Emission rates for VOCs are shown in Table 1 for the individual tests and in Table 2 for the composite profiles. The highest emission rates are observed from burning hardwoods in the wood stove (21.8 g/kg), and the softwoods combusted in the fireplace showed the lowest emission rates (5.8 g/kg) among nonsynthetic woods. The synthetic log emitted less VOC (1.2 g/kg) than the woods. The higher VOC emissions from wood stove as compared to fireplace can be attributed to higher combustion temperatures and a longer burn cycle. Contrary to the fine particle emission rates, moisture content had a noticeable effect on the emission rates of VOCs. The high-moisture wood stove tests yielded approximately 2–4 times more VOC than the next highest wood combustion test (Table 1).

Data regarding the impact of residential wood combustion VOC to ambient airsheds are currently sparse. Such data are important due to the presence of many compounds that are potentially toxic and/or highly reactive in ambient air. To estimate the contribution of some selected pollutants from wood combustion in an area impacted by wood smoke, emission inventory activity data for Washoe County (Reno), NV, in 1998 were obtained. On the basis of these data and the data obtained during the current study, it is estimated that 18.7 tons of benzene, 6.0 tons of formaldehyde, and 3.9 tons of 1,3-butadiene were emitted from residential wood combustion in Washoe County during 1998.

Carbon monoxide (CO) emissions, shown in Table 1 and Table 2 for the wood stove tests, averaged 129 ± 25 g/kg. CO emissions factors from these tests are comparable to those found in other emissions studies, as shown. Table 1 shows that hardwoods combusted with a low fuel load and high burn rate provided an enrichment in the carbon monoxide emission rate (178.6 g/kg).

Aliphatic and Olefin Hydrocarbons. Volatile and semivolatile hydrocarbons from C₂ to C₂₀ were quantified during

TABLE 2. Fine Particle and Gaseous Emission Rates from Softwood (Fireplace), Hardwood (Fireplace + Wood Stove), and Synthetic Logs (Fireplace)^w

	method	fireplace, softwood		fireplace, hardwood		wood stove, hardwood		synthetic log (mg/kg)	other ref (mg/kg)	
		mg/kg	median	mg/kg	median	mg/kg	median			
C2–C6 Hydrocarbons										
acetylene	can	312.97 ± 11.67	na	539.85 ± 41.09	na	1119.13 ± 59.04	na	nq		
alkanes										
ethane	can	468.51 ± 17.47	na	662.88 ± 181.79	na	1425.67 ± 977.85	na	nq	735(4)	
propane	can	107.43 ± 24.79	108.52	167.84 ± 57.70	157.08	155.06 ± 79.73	158.27	180.73 ± 4.01	331(13), ^j 45(13) ^k	
<i>n</i> -butane	can	17.36 ± 11.9	21.35	20.24 ± 19.60	23.42	1.67 ± 3.34	0.00	18.24 ± 0.81	55(13), ^j 6(13) ^k	
<i>n</i> -pentane	can	8.69 ± 0.84	8.88	11.85 ± 5.29	8.83	13.12 ± 7.81	14.12	7.90 ± 0.46		
<i>n</i> -hexane	can	6.01 ± 2.03	5.59	7.02 ± 3.77	5.01	12.30 ± 7.47	14.19	6.16 ± 0.35		
alkenes										
ethene	can	715.05 ± 26.66	na	1069.78 ± 319.90	na	2528.65 ± 424.92	na	nq	2245(4)	
propene	can	243.64 ± 49.35	230.51	346.20 ± 136.74	295.86	667.97 ± 439.24	589.47	30.38 ± 1.51	888(13), ^j 331(13) ^k	
1-butene/isobutene	can	84.13 ± 17.63	79.29	120.68 ± 50.68	101.07	148.85 ± 72.71	159.29	14.64 ± 0.99		
1,3-butadiene	can	62.63 ± 12.71	59.47	94.67 ± 40.80	73.51	196.58 ± 143.78	158.38	8.48 ± 0.58		
1-pentene	can	10.33 ± 1.04	10.42	15.19 ± 8.11	10.63	18.73 ± 11.64	18.98	7.49 ± 0.58		
1,3-pentadiene	can	7.35 ± 4.28	5.79	12.34 ± 7.36	9.84	0.00 ± 0.01	0.00	0.64 ± 0.06		
1-hexene	can	11.15 ± 2.69	11.03	18.55 ± 12.86	10.77	19.57 ± 19.11	19.26	10.22 ± 0.58		
1,3-cyclopentadiene	can	26.84 ± 9.15	29.39	27.60 ± 21.91	35.05	127.88 ± 115.59	80.90	0.93 ± 0.06		
C7–C20 Hydrocarbons										
alkanes										
<i>n</i> -heptane	can	3.71 ± 0.26	3.63	5.36 ± 3.23	3.51	4.60 ± 2.04	5.16	5.00 ± 0.29		
<i>n</i> -octane	can	2.54 ± 0.71	2.39	3.90 ± 3.00	1.94	14.94 ± 21.78	5.57	8.13 ± 1.05		
<i>n</i> -nonane	can	1.09 ± 0.26	1.09	2.31 ± 1.90	1.50	5.13 ± 3.56	4.27	0.00 ± 0.06		
<i>n</i> -decane	can	0.92 ± 0.26	0.84	2.10 ± 1.94	1.78	1.67 ± 0.71	1.82	2.73 ± 0.17		
<i>n</i> -undecane	can	2.27 ± 0.93	2.18	2.92 ± 2.57	4.59	1.29 ± 1.28	1.16	1.86 ± 0.29		
<i>n</i> -dodecane	can	0.93 ± 0.35	1.02	2.54 ± 2.47	3.19	2.18 ± 1.96	2.17	0.81 ± 0.23		
<i>n</i> -tridecane	Tenax	1.60 ± 2.27	na	1.45 ± 2.91	0.00	0.40 ± 3.23	0.09	0.51 ± 0.45		
<i>n</i> -tetradecane	Tenax	2.16 ± 1.19	na	0.00 ± 0.08	0.00	0.92 ± 1.85	0.14	0.89 ± 0.49		
<i>n</i> -pentadecane ^a	Tenax	4.71 ± 1.05	na	14.41 ± 12.49	13.14	1.82 ± 3.64	0.19	1.64 ± 0.37		
<i>n</i> -hexadecane	Tenax	0.73 ± 0.66	na	1.91 ± 2.02	1.36	0.11 ± 0.25	0.08	0.41 ± 0.38		
<i>n</i> -heptadecane	Tenax	0.94 ± 0.71	na	0.86 ± 0.75	0.58	0.11 ± 0.11	0.05	0.06 ± 0.12		
<i>n</i> -octadecane	Tenax	1.14 ± 0.75	na	1.33 ± 1.18	0.83	0.11 ± 0.30	0.06	0.76 ± 0.32		
<i>n</i> -nonadecane	Tenax	1.32 ± 0.74	na	1.89 ± 1.08	2.22	0.12 ± 0.28	0.10	0.46 ± 0.27	8.8(2) ^j	
<i>n</i> -eicosane	Tenax	1.04 ± 0.90	na	0.60 ± 0.60	0.49	0.13 ± 0.15	0.09	2.79 ± 1.73	21.7(2) ^j	
alkenes										
1-heptene	can	3.22 ± 0.77	3.09	5.15 ± 3.94	2.85	6.70 ± 4.45	6.92	3.20 ± 0.06		
1-octene	can	1.49 ± 0.52	1.35	0.43 ± 0.59	0.00	0.18 ± 0.35	0.00	7.49 ± 0.12		
1-nonene	can	0.49 ± 0.59	0.41	0.50 ± 0.39	0.44	0.00 ± 0.01	0.00	1.22 ± 0.06		
1-dodecene	can	1.06 ± 0.85	0.78	1.23 ± 1.06	0.91	0.77 ± 0.72	0.63	0.70 ± 0.12		
Oxygenated										
furans										
furan	can	132.52 ± 38.02	123.34	213.65 ± 162.75	157.86	208.49 ± 100.44	222.49	2.27 ± 0.17	225(13), ^j 133(13) ^k	
2-methylfuran	can	127.63 ± 51.36	135.24	209.86 ± 165.84	167.29	261.93 ± 169.61	279.50	3.89 ± 0.87	1076(13), ^j 39(13) ^k	
2,5-dimethylfuran	can	25.80 ± 13.44	24.83	36.32 ± 37.29	30.38	43.15 ± 22.83	47.82	0.23 ± 0.06	277(13), ^j 10(13) ^k	
2,4-dimethylfuran	can	2.71 ± 1.17	2.69	2.31 ± 1.78	1.87	2.89 ± 2.01	3.44	0.70 ± 0.06		
2-furaldehyde	Tenax	317.60 ± 258.17	na	445.33 ± 473.84	244.73	99.71 ± 80.13 ^b	na	4.21 ± 3.39	790(32)	
ketones										
glyoxal	DNP	424.32 ± 302.63	480.21	599.97 ± 470.71	573.12	172.68 ± 123.79	191.76	18.86 ± 6.54		
acetone (+propanal)	can	258.86 ± 69.74	256.31	426.10 ± 211.34	274.37	549.18 ± 463.84	391.63	106.66 ± 22.55		
3-buten-2-one	can	45.63 ± 9.32	46.61	58.76 ± 34.38	54.58	174.59 ± 119.24	174.23	2.32 ± 0.52		
butanone	can	80.40 ± 15.47	78.38	138.84 ± 44.19	133.28	172.71 ± 96.68	180.76	9.93 ± 0.87	335(13), ^j 35(13) ^k	
3-methyl-3-buten-2-one	can	52.20 ± 16.87	50.70	58.12 ± 43.68	51.52	80.27 ± 52.26	85.21	2.38 ± 1.22		

TABLE II (Continued)

	method	fireplace, softwood		fireplace, hardwood		wood stove, hardwood		synthetic log (mg/kg)	other ref (mg/kg)
		mg/kg	median	mg/kg	median	mg/kg	median		
Oxygenated									
alkanals									
formaldehyde	DNPH	113.32 ± 34.67	114.71	177.95 ± 53.03	164.51	245.58 ± 48.58	261.87	35.95 ± 4.95	100–700(14)
acetaldehyde	DNPH	301.03 ± 89.75	282.54	450.43 ± 87.57	425.01	360.46 ± 161.75	415.94	45.13 ± 4.39	30–600(14)
propanal	DNPH	80.22 ± 39.72	87.68	150.10 ± 82.55	139.44	96.44 ± 48.51	99.91	18.88 ± 5.50	100–300(14)
butanal	can	18.64 ± 1.26	18.31	22.48 ± 14.61	26.54	36.49 ± 26.56	36.29	9.06 ± 0.70	
pentanal	can	6.81 ± 2.40	6.37	6.26 ± 3.96	4.56	13.05 ± 9.92	12.05	6.74 ± 0.35	
octanal	can	8.87 ± 6.73	5.73	1.02 ± 0.82	0.65	1.55 ± 1.12	1.42	0.58 ± 0.12	
nonanal (+undecene)	can	7.85 ± 5.07	8.25	1.12 ± 0.94	0.88	2.27 ± 2.94	1.29	0.64 ± 0.12	2.97(2) ^f
alkenals									
acrolein	DNPH	46.90 ± 13.81	45.84	91.23 ± 34.72	81.94	45.54 ± 21.76	49.58	18.21 ± 2.90	20–103(14) ^g
butenal	can	19.62 ± 6.64	18.13	25.77 ± 14.42	28.58	22.52 ± 22.18	17.42	0.00 ± 0.06	
alcohols									
methanol (+methyl formate)	can	949.97 ± 344.63	862.59	2905.05 ± 1181.01	2624.75	3245.71 ± 2639.68	2849.57	0.00 ± 0.06	4300(32)
ethanol (+acn+acrolein)	can	208.91 ± 49.74	209.49	145.38	256.59	10525.75	619.08	114.38 ± 18.01	
other oxy compds		0.00 ± 0.00	0.00	0.00 ± 0.00	0.00	0.00 ± 0.00	0.00	0.00 ± 0.00	
ethyl acetate	can	3.72 ± 4.49	2.90	10.63 ± 3.25	8.83	1.17 ± 2.34	0.00	8.19 ± 0.06	
acetophenone	Tenax	64.95 ± 30.03	na	56.63 ± 48.87	53.56	10.90 ± 4.95 ^b	na	17.77 ± 8.07	
phenolic compds									
phenol	Tenax	157.58 ± 39.92	na	246.81 ± 174.46	246.92	142.37 ± 36.97 ^b	na	8.56 ± 2.17	110(32)
guaiacols									
guaiacol	F/P/X/P	77.30 ± 46.98	60.81	118.05 ± 21.65	128.07	21.69 ± 12.42	27.29	2.09 ± 0.28	180(32)
4-methylguaiacol	F/P/X/P	142.62 ± 58.96	141.48	76.10 ± 24.68	70.90	13.84 ± 8.10	16.80	3.20 ± 0.38	260(32)
4-ethylguaiacol	F/P/X/P	27.95 ± 9.99	27.03	22.40 ± 6.33	21.75	4.04 ± 2.35	4.80	0.85 ± 0.07	
4-allylguaiacol	F/P/X/P	22.57 ± 15.33	16.81	27.50 ± 10.87	24.80	1.55 ± 1.03	1.86	0.87 ± 0.34	76(32)
4-formylguaiacol	F/P/X/P	43.19 ± 20.94	33.71	47.31 ± 14.01	46.96	2.96 ± 2.33	3.69	7.09 ± 1.18	29.3(2), ^m 2.0(2) ⁿ
syringols									
syringol	F/P/X/P	1.57 ± 1.77	0.92	389.78 ± 132.08	405.20	45.69 ± 28.57	53.96	6.39 ± 0.47	1.1(2), ^m 10.8(2) ⁿ
4-methylsyringol	F/P/X/P	0.52 ± 0.22	0.43	244.28 ± 130.75	244.03	13.41 ± 9.26	18.46	3.42 ± 0.56	
4-ethylsyringol	F/P/X/P	1.75 ± 0.78	2.14	108.84 ± 55.98	138.47	5.19 ± 3.94	7.34	3.78 ± 0.69	
Halogenated									
chloromethane	can	27.67 ± 6.12	26.90	39.99 ± 15.14	42.47	21.06 ± 8.95	20.49	20.27 ± 1.68	
methylene chloride	can	176.19 ± 38.56	159.41	151.84 ± 47.08	172.74	2.12 ± 1.88	2.05	71.45 ± 4.82	
Resin/Terpenoid Compounds									
isoprene	can	38.74 ± 14.63	34.53	27.04 ± 13.39	24.80	43.25 ± 31.86	35.71	4.36 ± 0.23	
α-pinene	can	53.59 ± 24.05	54.65	4.01 ± 4.39	3.38	3.69 ± 2.55	4.10	5.81 ± 1.51	
β-pinene	can	59.68 ± 54.19	48.77	6.50 ± 8.12	2.81	16.56 ± 14.56	15.37	10.34 ± 0.41	
3-carene	Tenax	189.59 ± 56.36	na	18.15 ± 27.13	7.03	0.00 ± 0.01	0.00	0.00 ± 0.12	
limonene	Tenax	35.36 ± 8.71	na	2.89 ± 1.15	3.23	0.06 ± 0.13	0.00	0.37 ± 0.14	
sesquiterpene-e	Tenax	53.86 ± 25.86	na	0.00 ± 0.09	0.00	0.06 ± 0.06	0.07	0.00 ± 0.12	
Volatile Aromatic Compounds									
benzene	can	224.97 ± 40.25	229.02	312.15 ± 83.14	324.37	1189.68 ± 875.27	793.13	42.87 ± 2.90	988(13), ^j 951(13) ^k
toluene	can	130.62 ± 21.12	126.23	141.50 ± 44.03	138.52	320.00 ± 223.60	243.68	20.16 ± 1.05	373(13), ^j 241(13) ^k
<i>m</i> + <i>p</i> -xylene	can	49.63 ± 16.95	47.64	40.91 ± 12.22	39.56	71.78 ± 47.67	58.34	10.86 ± 1.10	
<i>o</i> -xylene	can	16.12 ± 4.19	15.02	19.15 ± 9.71	15.35	27.28 ± 16.80	24.55	7.09 ± 0.35	87(13), ^j 0(13) ^k
styrene (+heptanal)	can	40.17 ± 13.15	37.13	34.51 ± 22.29	37.67	117.21 ± 91.48	82.07	3.72 ± 0.35	
indan + indene									
indan	Tenax	0.12 ± 0.15	0.08	0.13 ± 0.19	0.00	0.49 ± 0.86	0.09	0.29 ± 0.06	
indene	Tenax	7.80 ± 7.77	6.23	1.06 ± 0.64	0.97	1.75 ± 1.69	1.36	0.29 ± 0.35	

TABLE II (Continued)

	method	fireplace, softwood		fireplace, hardwood		wood stove, hardwood		synthetic log (mg/kg)	other ref (mg/kg)
		mg/kg	median	mg/kg	median	mg/kg	median		
Gas-Phase PAH									
naphthalene	F/P/X/P	21.42 ± 5.13	22.57	54.62 ± 18.32	60.86	28.06 ± 19.06	34.96	20.58 ± 4.93	39(4), 84(6) ^o
2-menaphthalene	F/P/X/P	4.65 ± 0.84	4.76	12.90 ± 4.35	15.55	4.91 ± 3.33	6.32	2.03 ± 0.38	
1-menaphthalene	F/P/X/P	4.02 ± 0.71	4.14	10.33 ± 3.66	12.71	4.07 ± 2.77	5.23	2.28 ± 0.41	
biphenyl	F/P/X/P	1.18 ± 0.28	1.18	2.47 ± 0.98	2.63	2.43 ± 1.80	2.84	7.92 ± 1.87	
acenaphthylene	F/P/X/P	4.97 ± 1.03	5.35	8.67 ± 3.65	8.18	5.19 ± 3.65	6.40	6.70 ± 1.24	2(6), ^p 15(6) ^o
acenaphthene	F/P/X/P	0.41 ± 0.06	0.43	0.89 ± 0.29	0.77	0.52 ± 0.37	0.65	0.23 ± 0.04	5(4), 1.1(6) ^o
fluorene	F/P/X/P	2.15 ± 0.53	2.42	3.50 ± 1.08	3.09	1.66 ± 1.15	2.04	2.05 ± 0.31	2.1(6), ^p 6.7(6) ^o
Particle-Phase PAH									
phenanthrene	F/P/X/P	10.42 ± 2.74	10.17	16.59 ± 4.80	17.62	7.35 ± 5.00	9.12	7.62 ± 2.01	5.8(56) ^q
1-methyl-phenanthrene	F/P/X/P	2.30 ± 0.21	2.28	1.42 ± 0.49	1.61	0.26 ± 0.27	0.26	0.56 ± 0.05	0.19(56) ^q
1,7-dimethyl-phenanthrene	F/P/X/P	1.85 ± 0.17	1.86	1.09 ± 0.90	1.55	0.09 ± 0.06	0.12	0.62 ± 0.06	
retene ^c	F/P/X/P	1.79 ± 0.30	1.63	0.50 ± 0.22	0.54	0.02 ± 0.01	0.02	0.39 ± 0.04	0.68(2) ^m
pyrene	F/P/X/P	1.61 ± 0.24	1.65	3.39 ± 1.29	3.77	1.49 ± 1.03	1.82	2.99 ± 0.45	2.6(9), 2.7(56) ^q
indeno[123-cd]pyrene	F/P/X/P	0.07 ± 0.04	0.09	0.19 ± 0.11	0.18	0.08 ± 0.06	0.10	0.18 ± 0.02	0.42(56) ^k
benzo[e]pyrene	F/P/X/P	0.10 ± 0.01	0.10	0.25 ± 0.12	0.25	0.14 ± 0.10	0.16	0.26 ± 0.02	0.3(2), 6(4)
benzo[a]pyrene	F/P/X/P	0.15 ± 0.03	0.16	0.34 ± 0.19	0.30	0.20 ± 0.14	0.24	0.30 ± 0.04	0.6(56), ^q 2(4)
anthracene	F/P/X/P	2.50 ± 0.29	2.43	3.76 ± 1.21	3.46	1.43 ± 0.97	1.79	1.57 ± 0.18	0.82(56), ^q 3.1(6) ^r
fluoranthene	F/P/X/P	1.88 ± 0.29	1.93	3.99 ± 1.47	4.47	1.75 ± 1.21	2.14	3.26 ± 0.50	1.24(2), ^m 2.6(9)
benz[a]anthracene	F/P/X/P	0.31 ± 0.25	0.29	0.45 ± 0.34	0.48	0.56 ± 0.48	0.62	0.53 ± 0.38	0.16(6), ^p 0.63(6) ^o
chrysene	F/P/X/P	0.28 ± 0.04	0.28	0.61 ± 0.31	0.59	0.35 ± 0.25	0.43	0.36 ± 0.05	6(4), 0.26(6) ^p
benzo[<i>b+j+k</i>]fluorene	F/P/X/P	0.51 ± 0.08	0.51	1.05 ± 0.57	0.93	0.66 ± 0.48	0.79	0.94 ± 0.15	1.32(2), ^m 1.1(2) ⁱ
benzo[<i>ghi</i>]perylene	F/P/X/P	0.07 ± 0.06	0.09	0.22 ± 0.13	0.21	0.09 ± 0.06	0.11	0.24 ± 0.04	2(4), 1.3(9)
coronene	F/P/X/P	0.03 ± 0.02	0.03	0.09 ± 0.05	0.08	0.03 ± 0.02	0.03	0.09 ± 0.05	0.1(2), ⁱ 0.8–3(14)
Organic and Elemental Carbon									
organic C	TOR	3007.28 ± 333.54	3034.88	3579.95 ± 798.12	3486.60	2821.09 ± 1770.64	2861.28	1087.61 ± 108.75	2–20(14)
elemental C	TOR	774.15 ± 69.67	761.03	397.52 ± 117.67	395.15	356.33 ± 216.86	311.89	6578.23 ± 474.97	
Inorganic Species									
nitrate	IC	6.80 ± 1.42	6.48	9.94 ± 3.41	10.28	0.59 ± 3.83	0.09	2.24 ± 3.00	16.9(50), ^s 32.5(50) ^t
sulfate	IC	10.20 ± 3.33	8.90	27.51 ± 15.11	23.30	26.64 ± 14.62	26.59	0.00 ± 2.98	19.5(50), ^s 32.5(50) ^t
ammonium	CO	5.46 ± 1.58	5.12	5.05 ± 3.40	5.14	6.84 ± 8.32	3.31	9.44 ± 3.08	24.7(50), ^s 19.5(50) ^t
Elements									
chloride	IC	7.54 ± 3.69	5.56	10.07 ± 6.19	7.79	5.71 ± 3.79	6.02	0.00 ± 2.98	3.4(55), ^u 1.2(55) ^v
soluble potassium	AA	17.98 ± 7.38	16.65	67.40 ± 48.95	49.77	15.36 ± 11.85	12.01	1.42 ± 0.34	61.1(50), ^s 114.4(50) ^t
sodium	XRF	1.38 ± 2.16	1.17	1.08 ± 2.46	0.32	1.95 ± 6.79	1.37	6.89 ± 2.51	12.2(50), ^s 2.6(50) ^t
Summary of Emission Rates by Chemical Class^d									
alkanes(C3–C6)	can	192.7	200.1	239.7	223.1	238.1	242.7	292.7	
alkanes(C7–C12)	can	52.2	52.1	65.5	61.7	94.3	78.6	35.1	
alkenes(C3–C6)	can	531.1	509.8	757.0	648.4	1327.3	1161.5	88.5	
alkenes(C7–C12)	can	26.3	24.5	30.9	26.3	31.1	26.4	16.4	
furans	can	447.6	434.9	822.2	750.1	1024.2	1035.7	17.4	
carbonyls (C2–C9)	can	890.4	881.1	1293.3	1176.2	1794.0	1565.3	251.6	
other oxy	can	26.3	28.4	35.0	32.9	95.5	82.5	18.8	
alcohols	can	1201.4	1109.6	3388.5	3030.7	9175.0	3694.1	123.0	
resins/terpenoid	can	292.0	250.2	42.5	34.5	65.8	57.5	29.2	
aromatic (non-PAH)	can	653.0	629.7	658.6	670.4	2096.3	1488.7	108.9	
halogen	can	236.3	215.8	237.4	255.4	31.6	29.7	106.4	
identified VOC ^e	can	5769.0	5505.9	9448.0	8793.3	21796.8	13973.39	1235.6	12597 (13) ⁱ

TABLE II (Continued)

	method	fireplace, softwood		fireplace, hardwood		wood stove, hardwood		synthetic log (mg/kg)	other ref (mg/kg)
		mg/kg	median	mg/kg	median	mg/kg	median		
unidentified VOC ^r	can	245.5	216.6	389.6	362.1	893.9	650.6	161.9	
gaseous guaiacols ^g	F/P/X/P	247.9	229.3	216.6	204.0	39.6	47.6	6.1	
particulate guaiacols ^g	F/P/X/P	143.6	113.2	233.4	234.2	10.0	11.9	13.8	
syringols ^g	F/P/X/P	6.0	5.5	755.0	912.6	67.2	88.1	22.1	
gaseous PAHs ^h	F/P/X/P	43.4	45.2	108.3	121.0	54.5	68.9	48.6	
particulate PAHs ^h	F/P/X/P	36.5	36.2	59.1	64.0	20.2	25.2	26.6	
sum of elements		40.6	37.2	109.8	85.3	58.3	57.3	90.8	
Select Tenax Class Emission Rates (Not Included in Total VOC)									
phenol+methylphenols	Tenax	164.7	na	264.5	265.0	144.3	na	8.6	
sesquiterpenes	Tenax	140.7	na	4.0	4.1	0.1	0.07	0.1	
mass (g/kg)	gravimetry	5.14	4.92	5.66	5.95	3.96	4.02	8.27	
sum of particulate species ^(g/kg)	IR	4.43	4.39	4.78	4.76	3.81	3.75	7.98	13 (50), ^s 6.2 (50) ^t
CO (g/kg)		nq	nq	nq	nq	129.40	127.10	nq	148 (13), ^j 152 (13) ^k
isotopic C (% modern C)		100%	nq	100%	na	nq	nq	20%	

^a n-Pentadecane coelutes with some unidentified compounds. ^b Single wood stove profile from tenax. ^c Retene is 1-methyl-7-isopropylphenanthrene. ^d Emission summary includes all measured compounds (VOC summary from can data only); totals include compounds not shown in table. ^e VOC total based only on compounds quantified by the canister analysis. ^f Unidentified VOC includes unidentified hydrocarbons from the canister analysis. ^g Methoxylated phenol gas/particle distribution based on Hawthorne (28) work, where gas-phase guaiacols are guaiacol, methylguaiacol, and ethylguaiacol. ^h PAH gas/particle distribution assumed to occur at phenanthrene. ⁱ Sum of particle species includes particulate organics, elements (normalized for oxygenated species), elemental carbon, and organic carbon (normalized by multiplying by 1.2 (to compensate for oxy species) and by subtracting particulate species measured). ^j Ref 13, oak. ^k Ref 2, oak. ^l Ref 2, pine. ^m Ref 2, pine. ⁿ Ref 2, oak. ^o Ref 6, oak. ^p Ref 6, pine. ^q Ref 56, birch. ^r Ref 56, spruce. ^s Ref 50, pine. ^t Ref 50, hardwood. ^u Ref 55, softwood. ^v Ref 55, hardwood. ^w Abbreviations: can, canister; F/P/X/P, filter/PUF/XAD/PUF; XRF, X-ray fluorescence; TOR, thermal/optical reflectance; IC, ion chromatography; AA, atomic absorption spectroscopy; CO, colorimetry; IR, infrared; na, not applicable; nq, not quantified either because it was not seen or because it coeluted with another compound and could not be resolved.

this study. Most of the aliphatic and olefin hydrocarbon VOC is made up of the C₁–C₆ compounds. Wood-burning emissions contain higher proportions of ethene and alkadienes than typically found in other emission sources (36). Figure 2 shows that C₂ hydrocarbons constitute approximately 20% of the total measured VOC (C₂–C₁₂), with ethene (~12%) exceeding ethane and acetylene by approximately 4 and 5%, respectively. Acetylene, shown to be present in the wood combustion emissions, is often used as a marker species for mobile sources (37, 38) and has a strong influence on the attribution of motor vehicle exhaust in Chemical Mass Balance (CMB) calculations. Of the C₃–C₆ hydrocarbons, emissions of alkenes are at least twice as prevalent as alkanes (Table 2, Figure 2). The dienes, especially 1,3-butadiene and 1,3-cyclopentadiene, are enriched in wood combustion emissions and probably arise from the decomposition of terpenoids. In contrast, synthetic logs produce more alkanes than alkenes due to the presence of paraffin hydrocarbons in their structure.

Furans. Furans, which are formed from the decomposition of wood cellulose upon heating, constitute from 5 to 10% of the total C₂–C₁₂ VOC from softwood and hardwood emissions (Figure 2). Furan, 2-methylfuran, 2,5-dimethylfuran, and 2-furaldehyde are the most abundant of the furan species measured for all of the softwood and hardwood emissions. The structures of the pentoses commonly found in wood structure, xylose and arabinose, favor the production of furans with substituents at the 2 position.

Carbonyls. The carbonyl compounds, including ketones, alkanals, and alkenals, constitute between 10 and 18% of the C₂–C₁₂ VOC emissions from residential wood combustion (Figure 2). The C₄ ketones (butanone + 3-butene-2-one) and 3-methyl-3-butene-2-one are present in large amounts in softwood and hardwood emissions. These compounds most likely arise from the oxidation and decomposition of isoprene (2-methyl-1,3-butadiene) during combustion.

Lower molecular weight carbonyl compounds such as glyoxal, acetone, formaldehyde, acetaldehyde, and acrolein are the most abundant carbonyl species emitted. These compounds arise primarily from the combustion of cellulose (29). Table 2 shows that glyoxal emissions are the most abundant of these compounds, with 599.97 mg/kg being emitted from hardwoods combusted in the fireplace. Acrolein is emitted in the lowest amount, with hardwoods burned in the fireplace yielding 91.23 mg/kg. Carbonyl compound emissions from the synthetic log are lower as compared to the hardwoods and softwoods.

Alcohols. Alcohols, in particular methanol, constitute as much as 40% of the VOC emissions from residential wood combustion. Methanol has been postulated to originate primarily from the cleavage of methoxyl groups from lignin and secondarily from methyl uronic acid units in hemicelluloses (32, 39). These data (Table 2) are consistent with this proposed formation mechanism; methanol emissions are higher for hardwoods, which contain more methoxy groups in their lignin structure.

Lignin Breakdown Products (Methoxylated Phenols and Phenols). Synapyl and coniferyl alcohols consist of the syringol and guaiacol structural units, respectively (10, 26–28, 40–42). Guaiacol is the predominant precursor of softwood (gymnosperm) lignin. In the lignin polymer of hardwoods (angiosperms), structural units consist of both guaiacyl and syringyl types in roughly equal proportions. Thus, hardwood lignin shows a higher methoxyl content than that of coniferous woods (26).

Syringols and guaiacols are commonly found in wood-burning emissions (2, 3, 10, 27, 32, 34, 40–42). These wood lignin pyrolysis products are emitted in distinctive amounts and constitute as much as 21% of the total fine particle mass emissions during this study. As expected from variation in

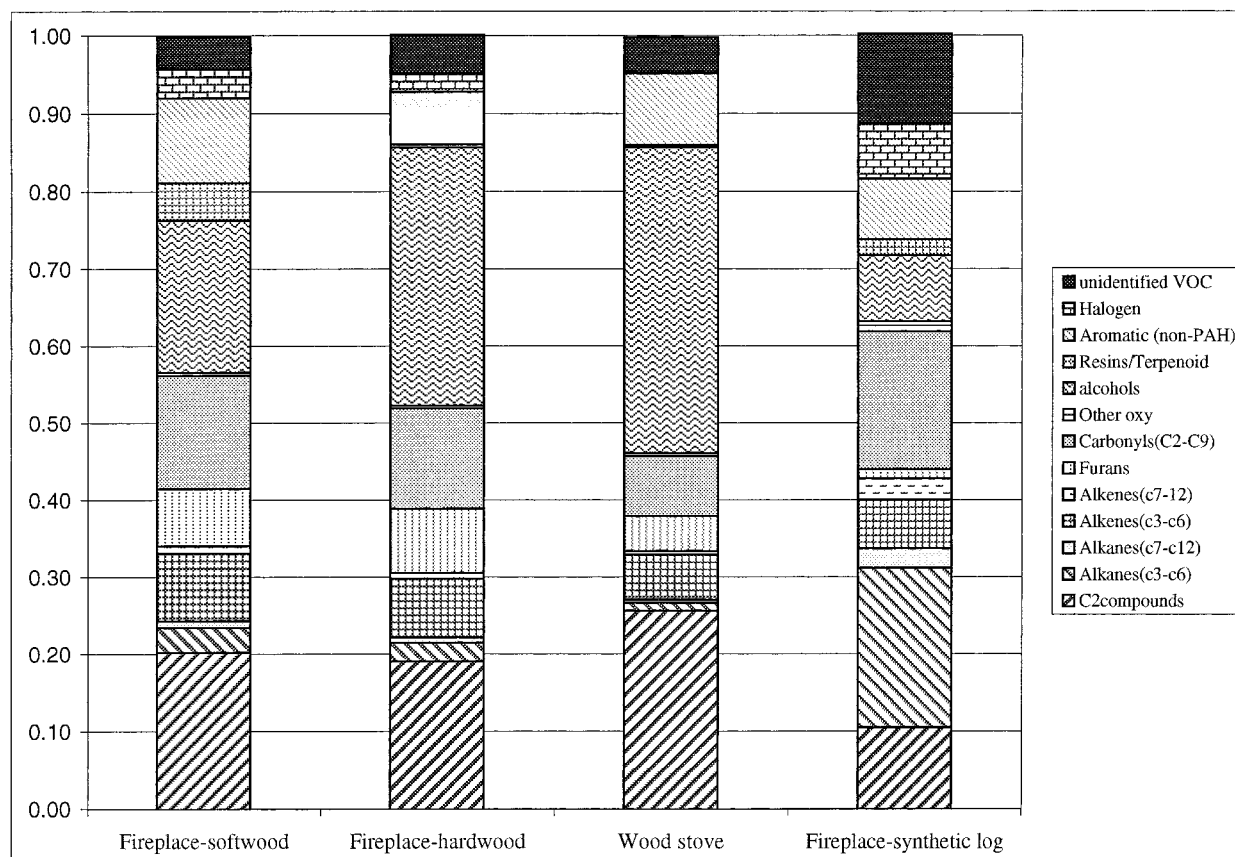


FIGURE 2. Weight fraction of volatile organic compounds (canister analysis only) from wood combustion source tests.

lignin composition, syringol abundances are larger in hardwood emissions than in softwood emissions. Syringol emission rates are 1.6 mg/kg for the fireplace/softwood composite, 389.8 mg/kg for the fireplace/hardwood composite, and 45.7 mg/kg for the wood stove composite.

Guaiacols are emitted in approximately equal amounts from both types of fuel in the fireplace, consistent with the results from Hawthorne et al. (28). The sum of the guaiacol compound emission rates are 391.5, 450.0, and 49.6 mg/kg for the fireplace/softwood composite, fireplace/hardwood composite, and wood stove composite, respectively.

Fireplace hardwood emissions are especially enriched with methoxylated phenols because hardwood emits both syringol and guaiacol compounds. Wood stove emissions contain less of the methoxylated phenols than the fireplace emissions. The methoxylated phenols appear to break down at the higher combustion temperatures encountered in the wood stove. The methoxylated phenols are also present in small amounts from the synthetic log emissions. This is not surprising because the synthetic log contains sawdust from real wood.

Phenol and the methyl phenols are also derived from the thermal decomposition of wood lignin. Table 2 shows that over 140 mg/kg of phenol is emitted from both hardwoods and softwoods in both appliances. Lower amounts of phenol are emitted during synthetic log combustion.

Halogenated Compounds. Edgerton et al. (43) used chloromethane to quantify wood smoke contributions to ambient particulate matter in Oregon. Table 2 shows that in addition to chloromethane, methylene chloride and methylchloroform are emitted from wood combustion. Methylene chloride is emitted at over 150 mg/kg for hardwoods and softwoods in the fireplace and at over 70 mg/kg from the synthetic log. The emissions of methylene chloride from the wood stove are considerably lower.

Resin Acids/Terpenoids. The principal biogenic emissions (not combustion related) from forest vegetation are isoprene for hardwoods and α/β -pinene for softwoods (44). As found in this study, isoprene is emitted in similar amounts from hardwoods and softwoods in both appliances. The terpenoids quantified during this study include the pinenes, carene, limonene, and a series of sesquiterpene isomers (shown in Supporting Information). These terpenoids are enriched in the softwood combustion emissions. Softwoods contain oleoresins that are composed of terpenoids and resin acids. These compounds are secreted by the resin-forming cells of the sapwood when the tree is wounded by scarifying, boring, etc. (30). The composition of these emissions depends on the genus and species of the softwood being burned. The Ponderosa and pinion pines tested during this study are especially enriched in 3-carene, emitting a composite average of 189.5 mg/kg.

Retene (1-methyl-7-isopropylphenanthrene) and 1,7-dimethylphenanthrene have been proposed as markers for wood burning emissions that are formed by combustion reactions of abietic and pimaric resin acids that are present in softwood (28, 46, 47). Retene, 1-methyl-phenanthrene, and the 1,7-dimethylphenanthrene isomers are more abundant in softwood as compared to hardwood (Table 2). Retene emission rates for the softwood fireplace composite are 4 times those of the hardwood fireplace composite and 90 times those of the wood stove hardwood composite. However, the emission rates of these compounds are low as compared to the methoxy phenols emission rates, and their use in apportionment studies are limited when their concentrations are at or below ambient detection limits (10, 11).

Volatile Aromatic Compounds. Residential wood combustion is a notable source of benzene, toluene, and the xylenes. Hardwood combusted in the wood stove emits over 1 g of benzene/kg of wood burned. Considerably more of the

aromatic compounds come from the wood stove combustion than other appliances. The high temperatures and long smoldering conditions encountered during a wood stove cycle favor the emission and formation of aromatic species.

There is a considerable difference in the amount of indan emitted from wood combustion as compared to indene. Indene is likely to be formed from the same pathway as benzofuran, which has a similar structure. The dehydration of and cyclization of cellulose could favor the formation of indene over indan.

Polycyclic Aromatic Hydrocarbons. Polycyclic aromatic hydrocarbons (PAH) are an important component of residential wood smoke emissions (9, 12, 13, 48–50). The fuel itself does not contain PAH, so these compounds must arise from pyrosynthesis (12). The presence of lignin, the only aromatic present in wood, has been shown to increase the formation rate of PAH during combustion (33).

For the purpose of this study, PAH are divided into two broad categories: gas phase and particle phase, even though large groups of the three–four-ring PAH are distributed between both gas and particle phases. Gas-phase PAH are defined as those that elute from a nonpolar capillary chromatographic column between naphthalene and phenanthrene (not including phenanthrene). Particle PAH include all PAH eluting between phenanthrene and coronene. The ratio of gas to particulate phase PAH is approximately 1:1 for softwood combusted in the fireplace, 2:1 for hardwood in the fireplace, and 3:1 for hardwood in the wood stove. The production of the more complex particle PAH, which are formed at elevated combustion temperatures, were expected to be enriched in the wood stove emissions. However, the slower burn cycle characteristic of wood stove combustion consists of a longer lower temperature smoldering condition that may favor the production of gas-phase PAH. The total PAH emission rate for fireplace/softwood and fireplace/hardwood combustion are 79.8 and 167.4 mg/kg, respectively. The average emission rate for total PAH compounds from wood stoves is 74.7 mg/kg. Similar to the results of Rogge et al. (2), synthetic logs were found to be a source of PAH emissions, yielding 75.2 mg/kg.

Khalil (48) reported that the predominate PAHs in wood smoke emission samples were acenaphthylene, naphthalene, anthracene, phenanthrene, benzo[a]pyrene (BaP), and benzo[e]pyrene (BeP). Two- and three-ring PAHs were responsible for 70% of the measured PAHs (49). Table 2 shows similar results from this study, with unsubstituted naphthalene, phenanthrene, and acenaphthylene being emitted in the largest amounts.

Organic and Elemental Carbon. The ratio of total organic carbon (OC) to total elemental carbon (EC) can be useful in distinguishing carbonaceous sources. The ratio of OC/EC for the softwood is 3.9, as compared to 9.0 for the hardwood burned in the fireplace and 7.9 in the wood stove.

Hildeman et al. (50) found that organic carbon was more prevalent in synthetic log emissions than elemental carbon, while this study shows the opposite. These differences are possibly a result of different burning conditions for the individual tests conducted. Differences may also result from the different analytical methods used for the analysis of carbon in these two studies. Chow et al. (24) described the importance of the analytical method used when defining the measured split between organic and elemental carbon.

Elements + Ions. Trace elements and ionic species such as water-soluble potassium, chloride, and sulfate are emitted from wood combustion (1). Turn et al. (51) showed that many of these species make up large amounts of the unburned inorganic portion of wood mass. As mentioned previously, these trace elements have been commonly used with the total organic and elemental carbon concentrations to apportion wood smoke in studies that did not utilize organic

chemical components. These species are limited as wood combustion markers, however, by their presence in other types of vegetative burning and meat cooking (10, 52).

Emissions of water-soluble potassium from wood combustion are highly variable, with hardwoods burned in the fireplace emitting 3–4 times more than softwoods or hardwoods in the wood stove. The synthetic log combustion emitted only 1.4 mg/kg dry fuel of water-soluble potassium during these tests (Table 2). Chlorine emissions range from 1.3 mg/kg in the synthetic log to 8.9 mg/kg from the fireplace/hardwood. Sulfate emissions are enriched in hardwood combustion in the wood stove (27.5 mg/kg), which emits on average almost 4 times more sulfate than softwood in the fireplace (10.2 mg/kg).

Isotopic Carbon. Isotopic carbon measurements of the $^{14}\text{C}/^{12}\text{C}$ abundance in ambient air and source samples show the fraction of modern versus fossil derived carbon. This isotopic carbon distribution has been used to apportion the contributions of vegetative burning (modern carbon) in ambient air. During this study, isotopic measurements were conducted for selected samples of hardwoods and softwoods and a synthetic log burned in the fireplace. The results for hardwood and softwood burning (Table 2) showed modern signatures, as expected. The synthetic log sample showed a mixed fossil and modern signature (Table 2) consistent with the wood chips/petroleum wax composition of this source.

Source Profiles. As mentioned previously, the primary objective of this study was to produce source profiles for wood combustion that can be used as input files in source attribution studies. For receptor modeling, methoxylated phenols and ions are useful for the attribution of fine particle emissions from wood combustion. Select PAH, such as retene, 1,7-dimethylphenanthrene, and 1-methylphenanthrene, can be useful but are limited to emissions mostly from softwoods and they are emitted in low amounts (<0.1% of fine particle mass) relative to the other species mentioned. Levoglucosan, a compound not reported in this study, is emitted in copious amounts from wood combustion and is a potentially useful compound for source apportionment (53). Among the volatile fraction, terpenoids, methyl chloride, furans, and the ketones can be useful for both hardwood and softwood apportionment. Caution must be exercised in using many of these compounds, however, due to their potentially short atmospheric lifetimes.

For the purposes of calculating weight fractions of semivolatiles and particle compounds for source profiles, a sum of particulate species is used as the denominator. This sum includes particulate organics, elements (normalized for oxygenated species by multiplying $1.89 \times \text{Al}$, $2.14 \times \text{Si}$, $1.65 \times \text{Cl}$, $1.4 \times \text{Ca}$, and $1.43 \times \text{Fe}$), elemental carbon, and organic carbon. Organic carbon results used for the sum of particulate species are adjusted by multiplying by 1.2 to account for unmeasured hydrogen + oxygen (54) and by subtracting the particle organic species measured.

The total VOC used as the denominator for calculating weight fraction of individual VOC (Figure 2) is a sum of all identified and unidentified compounds measured by the canister analysis. A standard protocol has not been established to define the denominator for VOC profiles used in source attribution studies. Fujita et al. (36) has proposed using the Photochemical Assessment Monitoring Stations (PAMS) list of compounds as the denominator. The PAMS species are emitted from most combustion sources and can account for 70–80% of the total ambient hydrocarbons in most urban locations (36).

Summary

This paper describes a comprehensive analytical effort to analyze the fine particle and gaseous emissions from residential wood combustion. Over 350 compounds and

elements were quantified for emissions of wood smoke from appliances, fuels, and burn conditions that have been shown in previous studies to effect both the composition and the emission rates from wood burning. During this study, the appliance used (fireplace versus wood stove) and the type of wood burned (hardwood, softwood, synthetic log) had the largest effect on the composition of emissions from wood combustion. Four distinct composite emission rate profiles were constructed to represent softwoods, hardwoods, and synthetic logs burned in a fireplace as well as hardwoods burned in a wood stove.

By creating source profiles according to guidelines described in this paper, these data may be used in source attribution studies for either fine particles or VOC. Until recently (57), wood combustion has not been included in VOC source apportionments. On the basis of the emission rates measured in this study, wood combustion should be included when evaluating the impact of combustion sources on VOC in ambient airsheds. In addition, the data presented here improve the current knowledge base for the types of pollutants emitted from wood combustion and their emission rates; especially hazardous air pollutants such as 1,3-butadiene, benzene, formaldehyde, and others that are not considered in current assessments of wood combustion emissions.

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