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Fine Particulate Matter (PM) and Organic Speciation of Fireplace Emissions

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This paper presents a summary of fireplace particle size and organic speciation data gathered to date in an ongoing project. Tests are being conducted in a residential wood combustion (RWC) laboratory on three factory-built fireplaces. RWC wood smoke particles <10 μm (PM10) consist primarily of a mixture of organic compounds that have condensed into droplets; therefore, the size distribution and total mass are influenced by temperature of the sample during its collection. During the series 1 tests (15 tests), the dilution tunnel used to cool and dilute the stack gases gave an average mixed gas temperature of 47.3 °C and an average dilution ratio of 4.3. Averages for the PM2.5 (particles <2.5 μm) and PM10 fractions were 74 and 84%, respectively. For the series 2 tests, the dilution tunnel was modified, reducing the average mixed gas temperatures to 33.8 °C and increasing the average dilution ratio to 11.0 in tests completed to date. PM2.5 and PM10 fractions were 83 and 91%, respectively. Since typical winter time mixed gas temperatures would usually be less than 10 °C, these size fraction results (even from the series 2 tests) probably represent the lower bound; the PM10 and PM2.5 size fractions might be higher at typical winter temperatures. The particles collected on the first stage (cutpoint ≈ 11.7 μm) were light gray and appeared to include inorganic ash. Particles collected on the remainder of the stages were black and appeared to be condensed organics because there was noticeable lateral bleeding of the collected materials into the filter substrate. Total particulate emission rates ranged from 10.3 to 58.4 g/h; corresponding emission factors ranged from 3.3 to 14.9 g/kg of dry wood burned. A wide range of Environmental Protection Agency (EPA) Method 8270 semivolatile organic compounds were found in the emissions; of the 17 target compounds quantified, major constituents are phenol, 2-methylphenol, 4-methylphenol, 2,4-dimethylphenol, and naphthalene.

Introduction

In 1995, EPA estimated that residential wood combustion (RWC), including fireplaces, accounted for 0.83% of PM10 emissions and 12% of air toxics (*1*). Based on very limited wood stove particle size data (*2*), it has been assumed that wood stove and fireplace emissions are 100% <2.5 μm. This has led to the assumption that RWC accounts for 4−5% of national PM2.5 emissions. Because of the uncertainty in these data, it was decided to undertake a sampling project to firm up the particle size data for RWC emissions and also look at composition as a function of particle size range. Even though the EPA's program addressed both wood stoves and fireplaces, this paper is focused on reporting the measurement of the particle size distribution of fireplace emissions. Also included are total particulate matter emission rates and the results of analyses for semivolatile organics in the emissions. This paper is an update of material presented previously (*3*).

Experimental Discussion

Appliances Tested. Three factory-built fireplaces were tested in the EPA/ARCADIS Wood Stove Laboratory. Two of them (A and B) were standard designs typical of those installed by builders in new homes. The third unit (C) was a low emission design incorporating secondary air tubes similar to those found in most current, EPA-certified noncatalytic wood stoves. The firebox on the low emission fireplace is smaller than in most of the conventional designs. It incorporates secondary air, injected through three perforated, horizontal tubes extending across the width of the firebox near its top. All of the fireplaces were supplied with glass doors and an adjustable damper for admitting outdoor (outside the dwelling) air into the firebox. Fireplace B also came equipped with a room air blower which circulated room air through channels between the firebox metal wall exterior and the insulated exterior surface wall.

Data from the series 1 tests, consisting of 14 test burns and a blank, and 2 test burns from the ongoing series 2 tests are reported in this paper. All tests basically followed the Washington State (WA) protocol (*4*). Three exceptions were as follows: (1) red oak cordwood was used for 12 series 1 tests and both series 2 tests, (2) the fireplaces rested on a weigh scale during tests to measure burn rate, and (3) particulates were collected following EPA Method 5G (*5*). Douglas fir dimension lumber (as specified in the WA protocol) was used for the two remaining series 1 tests: moisture content was 30% in one and 22% in the other. The partially seasoned oak cordwood used in series 1 tests had an average moisture content of 34.2%. Moisture content of the well seasoned oak cordwood for the two series 2 tests, run to date, averaged 16.4% on a dry basis. Moisture content was measured with a commercially available electrical conductance meter. Each fireplace was tested under various combinations of glass doors open and closed, with and without outdoor air and, if so equipped, with and without the room air circulating fan on. All test burns began with a cold start and included three full wood loads. Wood load weight was determined as a function of firebox area following the WA protocol.

Total Particulate Matter Sampling. Emission sampling began when the stack temperature reached 25 °C above ambient and stopped when the stack temperature had fallen to 25 °C above ambient after loading the third wood load. All emissions from the fireplace stack were collected in a dilution tunnel that meets EPA's Method 5G specification. Shown schematically in Figure 1, the dilution system was modified between series 1 and series 2 tests to allow the introduction of cooled, air-conditioned air into the tunnel immediately downstream of the inlet. In series 2 tests to date, the procedure has been to inject the maximum amount of cooled air while maintaining sufficient air flow through

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TABLE 1. Fireplace EPA Method 5H Particulate Matter, Stack CO and CO_2, and XAD-2 Semivolatile Organic Test Results

test no./ fireplace code	dilution tunnel temp, °C	wood moisture, % H_2O dry basis	dry burn rate, kg/h	wet wood weight,[a] kg	method 5H equivalent[b]		stack gases,[b] g/kg dry wood		XAD-2 semivolatile organics,[b] g/kg dry wood
					g/h	g/kg dry wood	CO	CO_2	
1/A	43.9	34.2	3.81	27.0	56.13	14.74	109.9	2276.0	no data
2/A	40.3	34.2	3.62	27.0	53.80	14.86	123.8	2132.0	5.2
3/A	41.9	34.2	3.66	27.7	51.56	14.08	100.2	2178.0	8.3
4/A	43.3	34.2	3.85	27.4	56.13	14.56	110.1	2224.0	7.9
5/A	46.9	34.2	4.60	27.2	43.55	9.46	102.0	2244.0	4.2
6/B	50.3	34.2	4.44	34.1	42.61	9.60	87.5	1894.0	3.2
7/B	49.4	34.2	4.18	34.2	39.90	9.54	87.4	1922.0	1.1
8/B	54.2	34.2	4.96	35.4	42.20	8.50	93.7	2100.0	2.5
9/B	50.5	34.2	4.71	35.2	59.05	12.54	91.4	2096.0	0.9
10/B[c]	19.5				1.62	0.39	3.2	284.5	0.1
11/C	37.2	34.2	3.22	22.2	32.82	10.19	101.6	2071.0	5.9
12/C	42.1	34.2	3.20	21.0	21.25	6.63	87.6	2268.0	3.9
13/C	43.9	34.2	3.30	21.2	10.75	3.26	66.6	2608.0	2.4
14/B[d]	48.8	30.0	5.00	23.4	51.46	10.29	80.3	1789.0	3.3
15/B[d]	69.1	22.0	6.14	27.6	26.01	4.24	66.4	2431.0	ANC[f]
16/B[c]	32.4	17.4	4.87	34.6	40.00	8.21	72.1	2500.0	ANC
17/B[c]	35.1	15.4	7.29	34.2	26.88	3.69	46.2	2122.0	ANC

[a] Total wood burned for the test. Dry wood weight = wet wood weight ÷ [(% H_2O/100) +1]. [b] Values have been corrected for the blank values shown for Test 10. [c] The average burn rate for all tests (4.17 kg/h) was used to determine the g/kg blank value (g/h ÷ kg/h = g/kg) for Test 10. [d] Doublas fir 4 × 4s used for these tests. All other tests used split red oak cordwood. [e] Series 2 tests. Tests 1−15 constitute series 1 tests. [f] ANC (analysis not completed).

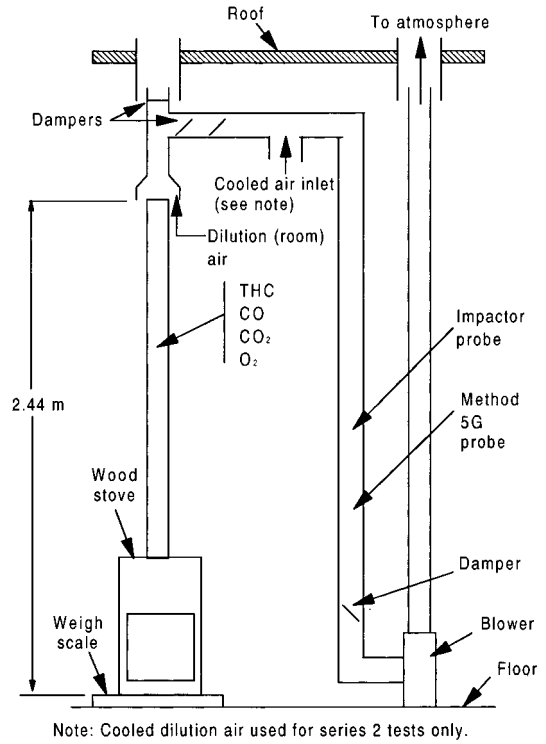

FIGURE 1. Typical fireplace test setup.

the tunnel inlet to entrap all fireplace emissions. The cooled air flowrate is not controlled during a test. The resulting mixed gas temperature will vary with the fireplace chimney flow and temperature. An improved dilution tunnel is currently under construction to allow for more accurate simulation of real-time conditions (such as, approaching mixed gas temperature of 10 °C) and to lend itself to a full characterization for particle loss.

As noted above, total particulate emission sampling followed EPA Method 5G. A continuous gas sample was extracted from the stack at the 2.44 m height and sent to a battery of analyzers to determine concentrations of carbon monoxide (CO), carbon dioxide (CO_2), and oxygen (O_2). Concentrations of these gases, stack temperature, and wood weight were recorded automatically at 1 min intervals throughout each test.

Particle Sizing. Particle size samples were collected with an Andersen Mark III 8-stage impactor with backup filter. The sampler was operated at 12−20 *l*pm, depending on the actual stack velocity; nozzle size was selected to give isokinetic conditions. The sampler was located inside the dilution tunnel aligned with the centerline, upstream of the Method 5G sample probe. Straight nozzles were used to preclude any size fractionation before the first impactor stage. The impactor's temperature was allowed to match that of the dilution tunnel gas. Prior to a test, impactor substrates were desiccated, weighed, and loaded into the clean sampler body. At the conclusion of all but Test 1, the entire impactor was placed in the desiccator for 24 h before disassembling it to recover the substrates. The recovered substrates were returned to the desiccator and allowed to reach constant weight before final weighing. Test 1 substrates were recovered at the end of the test. They were stuck to the supports and were destroyed in the recovery process. The Andersen sampler was not operated during the blank run.

Semivolatile Organic Sampling. A module containing an organic adsorbent, XAD-2, was placed immediately after the Method 5G filters to collect semivolatile organics. After exposure, the XAD-2 modules were kept in a freezer until they could be extracted. The resin was placed in a glass thimble which was then placed in a Soxhlet extractor. Extraction with dichloromethane continued for 16 h. The extracts were reduced to about 15 mR using a rotary evaporator and then concentrated to 5 mL with a water bath and dry nitrogen blowdown. Semivolatile organics are defined, in this case, as all organic compounds with boiling points between 100 and 300 °C. Sometimes referred to as total chromatographable organics (TCOs), this range includes all organics with carbon numbers from 8 through 16. Total semivolatile organics were determined by injecting an aliquot onto a gas chromatograph (GC) equipped with a flame ionization detector (FID) following EPA Air Pollution Prevention and Control Division standard methods (*6*). In a separate analysis, individual target compounds were determined by injecting an aliquot of each sample onto a Hewlett-Packard (HP) 5970 GC equipped with a HP 5970 mass

TABLE 2. Fireplace Test Configurations

test no./ fireplace code	outdoor air damper	glass doors	room air blower
1/A	closed	closed	NA
2/A	closed	closed	NA
3/A	closed	open	NA
4/A	closed	open	NA
5/A	open	closed	NA
6/B	open	closed	off
7/B	closed	closed	off
8/B	closed	closed	off
9/B	closed	open	on
10/B	NA	NA	NA
11/C	closed	open	NA
12/C	closed	open	NA
13/C	open	closed	NA
14/B	closed	closed	off
15/B	closed	closed	off
16/B[a]	closed	open	off
17/B[a]	closed	open	off

[a] Series 2 tests. Tests 1−15 constitute series 1 tests.

TABLE 3. Fireplace Particle Size Data

test no./fireplace code	% < than 2.5 μm	% < than 10 μm
1/A	sample lost	
2/A	81	89
3/A	84	91
4/A	85	92
5/A	77	91
6/B	84	91
7/B	77	81
8/B	65	66
9/B	85	91
10/blank	no data	no data
11/C	76	82
12/C	60	82
13/C	64	78
14/B	80	89
15/B	46	70
16/B	84	91
17/B	82	91
fireplace A average	82	91
fireplace B average	75	84
fireplace C average	66	81
wet wood average	76	85
seasoned wood average	71	84
overall series 1 average	74	84
overall series 2 average	83	91

spectrographic detector (MSD). Calibration of the GC/MSD used an EPA Method 8270 semivolatile working standard prepared by Radian Corporation. Quantification employed a five-point calibration curve.

Results

Total Particulate Matter. Each test ran for about 8 h. No filter changes during tests were required for either the total particulate or particle size sampling trains. In the WA fireplace test method, particulate emissions are reported in EPA Method 5H equivalents. The conversion equation used in this paper is (*7*)

$$\text{Method 5H} = 1.619(5\text{G})^{0.905} \quad (1)$$

The blank-corrected Method 5G particulate results from these tests, converted to their 5H equivalents with eq 1, are presented in Table 1, and the fireplace test configurations are presented in Table 2. Five observations of note are the following: only fireplace C would meet the WA standard of

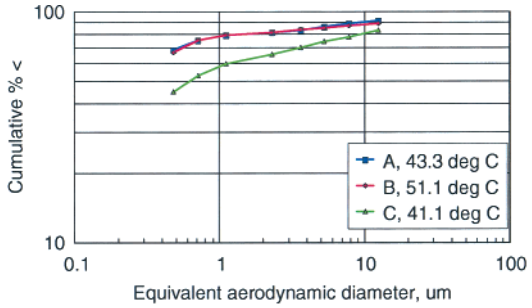

FIGURE 2. Effect of fireplace design on particle size distribution using wet oak fuel. Average dilution tunnel temperature shown in °C.

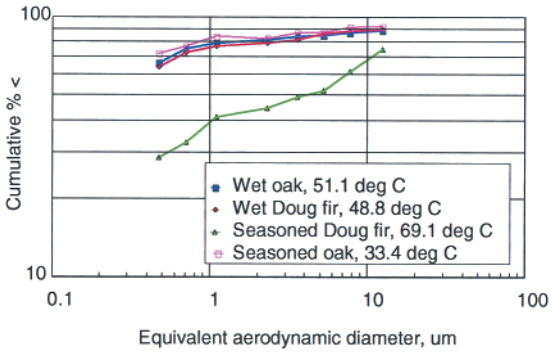

FIGURE 3. Particle size distribution from fireplace B burning wet and seasoned oak and Douglas fir. Average dilution tunnel temperature shown in °C.

≤7.5 g/kg while burning relatively wet (moisture > 30%) wood but then only in two out of three tests; fireplace B, burning normally seasoned wood, met the ≤7.5 g/kg WA standard in Tests 15 and 17 but not in Test 16; Tests 7 and 14, burning relatively wet oak and Douglas fir, respectively, gave comparable results, suggesting that wood species may not have a great effect on emissions; fireplace test configuration did not impact emissions significantly; and fireplace A had the highest emission factor.

Also included in Table 1 are the emission factors for CO, CO_2, and total semivolatile organics collected in the XAD-2 resin. The particulate matter and CO emission factors measured here are generally in the midrange of the values published by Dasch (*8*), Hall and DeAngelis (*9*), and Cooper (*10*), who investigated emissions from three fireplaces burning several wood species, including hardwood and softwood. Total semivolatile organics determinations for tests 15−17 have not been completed, although analysis for individual target compounds has been completed on Test 15.

Particle Sizing. Particle size results are presented in Table 3 and Figures 2 and 3. As noted previously, the particle size data from Test 1 were lost; no particle size samples were taken during the blank test (Test 10). Test 8 particle size results look spurious, especially the PM10 percentage. The overall averages for the relatively wet wood were 76.5 and 85.2% for the PM2.5 and PM10 fractions, respectively. Fireplaces A and B produced about the same size distribution when burning partially seasoned cordwood, especially if the Test 8 PM10 value is ignored (Figure 2). Fireplace C, the clean burning design, produced smaller fractions of PM2.5 and PM10 burning the same wood as fireplace A and B (Figure 2). Test 15, the one seasoned Douglas fir test burned in fireplace B, produced even smaller fractions of PM2.5 and PM10; the <2.5 μm fraction was only 46%, but note the higher dilution tunnel temperature (Figure 3). In contrast, Tests 16 and 17, burning well seasoned oak, averaged 83 and 91%,

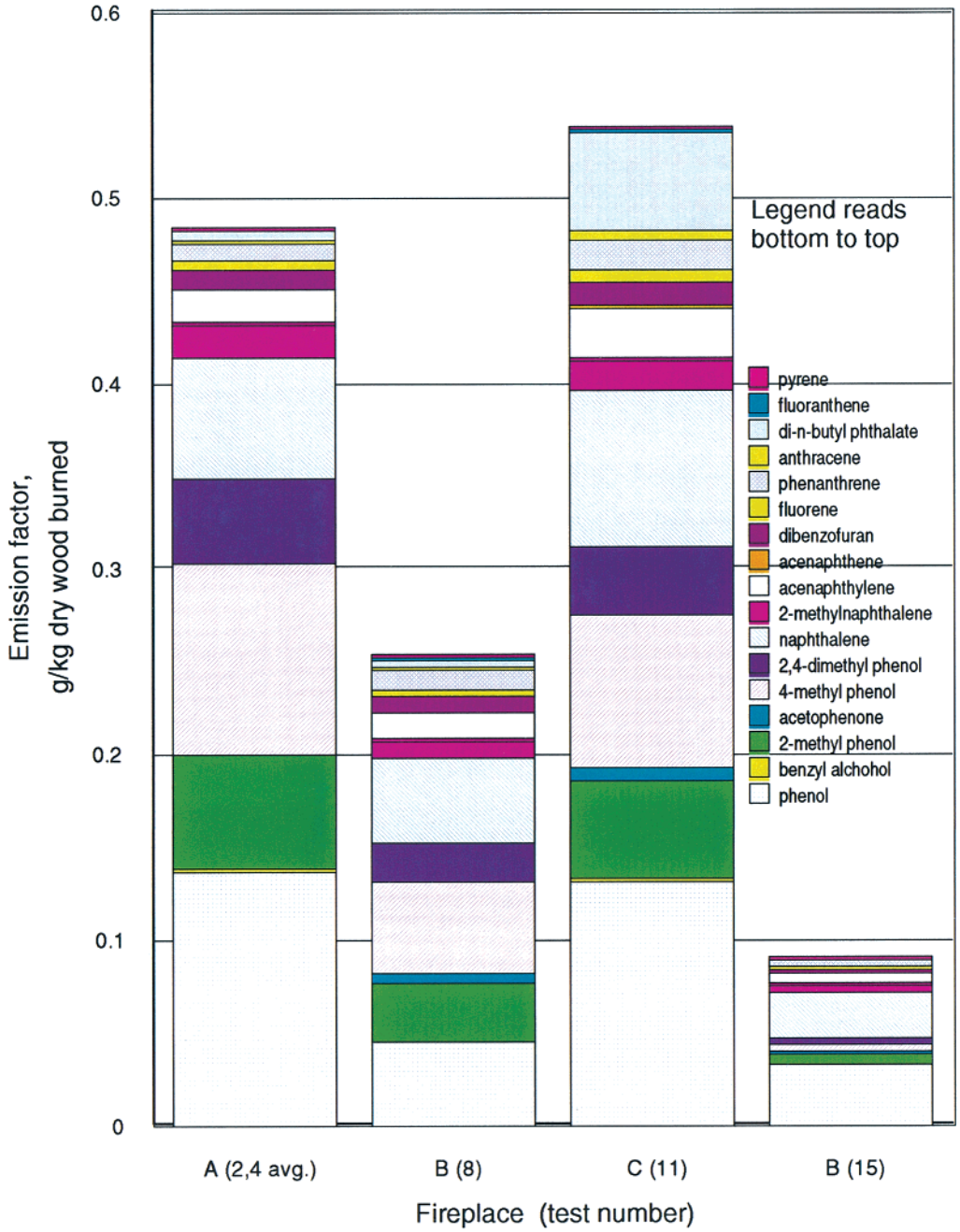

FIGURE 4. Emission factors for target semivolatile organic compounds (Table 4) found in the XAD-2 resin catches from selected emission tests on three factory-built fireplaces. Wet oak (34.2% moisture) cordwood used for all tests except seasoned Douglas fir (22% moisture) 4 × 4s used in Test 15.

respectively, when the dilution tunnel gas temperature was much lower (Figure 3).

The three variables, combustion efficiency, wood moisture, and dilution tunnel gas temperature, are interrelated and appear to have affected the particle size distribution. The CO_2 to CO ratio is a relative measure of combustion efficiency: a higher ratio means a higher efficiency. The clean-burning design (fireplace C) burning relatively wet oak, with an average CO_2/CO ratio of 28.5, generally produced less PM and a smaller PM2.5 fraction compared to fireplaces A (CO_2/CO = 20.4) and B (CO_2/CO = 22.2) burning the same wood. Fireplace B, burning drier wood (and with a considerably higher CO_2/CO ratio of 40.3 compared-to-a ratio of only 22.2 burning wetter wood), produced even less fine PM on Douglas fir but the same on seasoned oak as when burning wetter oak. Note that, compared to fireplaces A and B burning wetter wood, fireplace C produced less fine PM on the same fuel, again suggesting a combustion efficiency effect. Combustion efficiency seems to be more important than wood moisture alone, but it is not possible to separate the two effects.

The third variable, average dilution tunnel temperature, excluding Test 15, ranged from 37.2 to 54.2 °C (Table 1) with an overall series 1 tests average of 45.6 °C. The dilution tunnel temperature for Test 15, burning seasoned Douglas fir, averaged 69.1 °C. In series 2, it was 32.4 and 35.1 °C for the two tests run so far. As stated earlier, the impactor train was placed in the dilution tunnel and thus collected particles at the dilution tunnel temperature. RWC particulates (especially the fine fraction) are thought to be dominated by condensed organics. At the conclusion of the series 1 tests, it was hypothesized that the much smaller percent of the fine fraction collected while burning seasoned wood (Test 15) was due at least in part to the fact that less of the organics had condensed at the higher dilution tunnel temperature.

TABLE 4. Target and Other Semivolatile Organic Compounds Quantified/Identified in the Samples Collected on XAD-2 Resin

target (quantified) compounds	tentatively identified compounds
phenol	2-furancarboxaldehyde
benzyl alcohol	2-methylfuran
2-methylphenol	2-furanmethanol
acetophenone	2-butanone
4-methylphenol	ethylbenzene
2,4-dimethylphenol	1-(acetyloxy)-2-propanone
naphthalene[a]	xylene
2-methylnaphthalene	1,2-dimethylbenzene
acenaphthylene[a]	phenylthyne
acenaphthene[a]	styrene
dibenzofuran	2,5-hexanedione
fluorene[a]	pinene
phenanthrene[a]	2-methyl-3-pentanone
anthracene[a]	1-(acetoxy)−2-butanone
di-*n*-butyl phthalate	5-methyl-2-furancarboxaldehyde
fluoranthene[a]	benzaldehyde
pyrene[a]	benzofuran
	limonene
	indene
	2-methoxyphenol
	2-ethyl-phenol
	2-methoxy-4-methyl-phenol
	3-methoxy-1,2-benzenediol
	4-ethyl-2-methoxyphenol
	2,6-dimethoxy-phenol
	2-methoxy-4-(1-propenyl)phenol
	2,6-dimethoxyphenol
	1-methylnaphthalene
	bis(2-ethylhexyl)hexadenoic acid

[a] Included in the Clean Air Act (as amended) Section 112(c)(6) list of 16 POMs.

TABLE 5. Comparison of POM Emissions[a] from These Tests and Literature Values[b]

	these tests		literature (*11*)		
	wet oak	seasoned fir	seasoned oak (*9*)	green pine (*9*)	unknown (*10*)
sum of eight POMs common to both lists	0.106	0.037	0.0174	0.0184	0.0279

[a] In g/kg dry wood burned. [b] See Table 4 footnote *a*.

This hypothesis seems to be borne out by the results from Tests 16 and 17. Burning seasoned oak, the size fractions (PM2.5 = 83%, PM10 = 91%) are as high as or higher than those measured when burning wetter fuel (Figure 3 and Table 3). This is especially significant when considering that Tests 16 and 17 realized significantly higher combustion efficiency as evidenced by the reduction in CO causing the CO_2/CO ratio to nearly double (Table 1). Also note, however, that fireplace C produced a smaller PM2.5 fraction compared to A and B when burning the wetter wood, although the average dilution tunnel temperature was 41.1 °C, compared to 43.3 °C for all fireplace A tests and 51.1 °C for all fireplace B tests burning wetter oak.

Thus, of the three variables, it appears that sample collection temperature exerts the major effect on particulate size distribution. Combustion efficiency, per se, does not exert a major effect on particulate size distribution, but design modifications to lower emissions do, based on these very limited data. Future tests burning seasoned wood should shed more light on the particle size distribution issue.

Visual observation of the substrates from the individual impactor stages from all tests showed a consistent color trend. The first stage substrate, cutpoint ≈ 11.7 μm, was medium gray. The rest of the stage substrates were very dark gray to black, and there was some degree of bleeding into the substrate, indicating that at least some of the collected material was liquid drops.

Semivolatile Organics. XAD-2 samples from Tests 2, 4, 8, 11, and 15 were selected for GC/MS analysis. The results for the target compounds are shown in Figure 4. Note that the sums of the target compounds typically represent less than 10% of the total semivolatile organics collected in the resin shown in Table 1. The target compounds and other identified compounds quantified are listed in Table 4. Fireplace B burning relatively wet oak (34% moisture content, dry basis) produced less semivolatile emissions than did fireplace A burning similar wood. Surprisingly, the low emission fireplace C produced about the same quantity of semivolatile emissions as did fireplace A on the same fuel. Fireplace B's emissions dropped significantly when burning 20% moisture content Douglas fir. These data seem to indicate again that wood moisture content plays a major role in determining the quantity of emissions produced.

The target compounds included eight polycyclic organic matter (POM) compounds also quantified in the literature (*11*) (Table 4). A comparison of the sum of these eight POMs from the tests reported here and those in the literature is presented in Table 5. The data reported here for high moisture oak are nearly an order of magnitude higher than the literature values for seasoned oak and green pine. Seasoned fir produced about twice the emissions compared to green pine and about the same as for the unknown wood. Reference *11* also reports values for wood stoves; cordwood burning stoves range from 0.171 to 0.335 g/kg for the sum of the same eight POMs. These data compare favorably with the results from partially seasoned oak reported here.

Discussion

A total of 16 tests and a blank were run on three factory-built fireplaces. Twelve of these tests were conducted burning partially seasoned oak cordwood. Two burned well seasoned oak cordwood. Two tests burned air-dried Douglas fir dimension lumber: one test was with partially seasoned wood and the other with normally seasoned wood. The two conventional design fireplaces, currently certified to WA's 7.5 g/kg standard, exceeded this level while burning higher moisture content wood (both oak and Douglas fir). The clean burning design fireplace burning higher moisture wood and the conventional fireplace burning seasoned Douglas fir lumber both met that standard. The conventional fireplace also met the standard on one of two tests burning well seasoned oak cordwood.

Particle size distribution also seemed to be affected by (1) wood moisture content as it affects combustion efficiency and (2) the gas temperature at which the sample was collected. The PM2.5 size fraction varied from as high as 85% while burning wetter wood to 46% burning seasoned Douglas fir lumber, but the dilution tunnel temperature was significantly higher in the latter case. After modifying the dilution tunnel to reduce the mixed gas temperature, two tests were performed burning well seasoned oak cordwood, with a resultant average PM2.5 fraction of 83%. This indicates that the temperature at which the particle size sample is collected has a major impact on the measured distribution. Future testing will focus on gathering additional data at the lower dilution tunnel mixed gas temperature to better simulate field operation.

Literature values for the sum of the eight POMs common to these tests seem low, compared to data reported here, the latter being similar to wood stove emission levels. POM analyses will continue, including extraction and analysis of filter catches as well as the XAD-2 resin catches. These data, and data collected on several wood stoves, will be reported at a later date.

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