EMISSIONS FROM COAL-FIRED RESIDENTIAL COMBUSTION EQUIPMENT

by

Thomas W. Hughes and Darryl G. DeAngelis
Monsanto Research Corporation
Dayton, Ohio 45407

ABSTRACT

This paper presents the results of a study to quantify criteria pollutants and characterize other atmospheric emissions from coal-fired residential heating equipment. Flue gas was sampled from a warm air furnace and a hot water boiler which burned three western coals. Tests were conducted with the stokers operating on a 20-minute ON/40-minute OFF cycle, corresponding to high- and low-fire conditions in the fuel bed. Variations in coal composition and type of heating equipment both influenced emission rates, and the OFF portion of the heating cycle contributed significantly to total emissions. Combustion efficiencies for coal-fired residential heating equipment were lower than for larger coal-fired systems (e.g., utility boilers), as evidenced by the higher emission levels for CO and organic species, including POM's. In contrast to previous estimates, particulate emissions were not a function of the coal ash content, but did correlate with the coal volatile content. The particulate composition was primarily carbon, indicating that the particles were not formed from coal ash but from carbonaceous material volatilized during combustion.

INTRODUCTION

The majority of homes within the United States are now heated by either natural gas (56%) or fuel oil (24%). These fuels have traditionally been inexpensive, easy to obtain, clean, and simple to burn in home furnaces. However, as natural gas and oil reserves diminish, it has become apparent that alternate energy sources must be developed for the residential sector. One candidate for a replacement fuel is coal.

Coal is the nation's most plentiful fuel resource, and in the past it was the predominant home heating fuel. Recent advances in furnace design have made coal-fired residential furnaces easier to operate and nearly automatic. Consequently, the use of coal for home heating is attractive both economically and technically, and there are indications in some regions of the country of a trend toward increased home heating with coal.

One major drawback to residential coal combustion is an increased level of air pollution. As a result, both regional and national EPA officials have become concerned over the potential environmental impact from increased residential coal combustion. The major problem confronting the EPA has been the absence of an adequate data base upon which to make policy decisions. Until these emissions are adequately characterized, the EPA will lack sufficient information to formulate policies regarding an increased usage of coal in the residential sector.

The objective of this project was to assemble an emissions data base for coal-fired residential combustion. A stoker-fed boiler and a stoker-fed warm-air furnace were tested while burning western coals. Exhaust gases were measured for particulates, sulfur dioxide (SO_2), nitrogen oxides (NOx), carbon monoxide (CO), organic species including polynuclear organic materials (POM's), and individual elements.

Experimental Methods

Two different residential coal-fired heating units were tested burning three different coal types. Two of the coals were tested at three ash levels. Standard and modified sampling and analysis methods were used to measure emissions.

Heating Equipment

Two coal-fired heating units were selected for testing in this program: 1) a 200,000 Btu/hr hot-water boiler and 2) a 200,000 Btu/hr warm air furnace. Each unit was coupled with an underfeed stoker. The boiler was coupled with a stoker of a

more traditional design while the warm-air furnace stoker employed a mechanism to break up clinkers in the fuel bed. In addition to feeding coal to the burner, the stoker provided support units such as the retort, windbox, and combustion air supply.

During normal use the stoker feed screw and blower are activated by a room thermostat, a limit switch, and a hold-fire timing relay. For this study, however, it was necessary to maintain a constant combustion cycle in order to obtain consistent, reproducible stack gas samples. A timer was employed to override the above control devices by activating the stoker according to a predetermined 20-min ON/40-min OFF cycle. During the ON period combustion air is blown through the fuel bed to maintain a hot fire. During the OFF period residual coal in the fuel bed continues to burn, although at a much slower rate, maintained by natural draft from air leaks and vents. The draft is kept at a minimum by a barometric damper in the exhaust stack.

A previous study demonstrated that the combustion cycle was a significant factor influencing emission rates from coal-fired residential combustion units.[1] For this reason emissions were quantified during each cycle segment of the warm air furnace operation. Because of program constraints, emissions from the hot water boiler were measured for the total heating cycle or in some cases just for the ON position of the cycle.

Feed Coal

Heating units in this study were fired with 3 western coals designated as test coals A, B, and C. These coals were a high volatile B bituminous coal (Coal A), a high volatile C bituminous coal (Coal B), and a subbituminous B coal (Coal C). Coals A and B were purchased in two fractions, high ash (unwashed) and low ash (washed). Equal portions of each fraction were combined to generate a medium ash coal thereby giving three ash levels of each coal.

Coal quality in general was poor with excessive coal fines and rocks present in the shipment as received. Rocks could not be eliminated from the feed coal, but coal fines were avoided because they settled to the bottom of the shipping containers. Poor coal quality is common for coal available to the residential sector.

Sampling and Analysis Methods

Flue gas constituents were collected and measured using a variety of standard EPA procedures[2] or acceptable modifications and alternatives.

Particulates and Condensable Organic Matter

The procedure and equipment used for quantitative particulate collection met specifications outlined in Method 5 of the Federal Register.[2] A preliminary velocity traverse indicated a flat velocity profile across the stack. Because of the small stack diameter (8 in.), single-point sampling was used to determine the mass emission rates with the probe tip placed in the center of the exhaust stack.

Mass emissions from the boiler were collected over the entire heating cycle by a single Method 5 train. Mass emissions from the furnace were determined by using two Method 5 trains, one during the ON segment and the other during the OFF segment of the heating cycle.

Determination of condensable organic material was made from the back half portion of the sampling train. The back half of the train consists of water-filled impingers that collect most materials passing through the front half filter. Examination of the back half catch during these tests showed that it was condensable organic material, and it is reported separately in this way in the results section.

Sulfur and Nitrogen Oxides

Sulfur oxides and nitrogen oxides were collected and analyzed using equipment and procedures outlined in Method 6 and Method 7 respectively of the Federal Register.[2] Boiler samples were collected during the ON portion of the heating cycle while furnace samples were collected for both the ON and OFF segments.

Carbon Monoxide, Carbon Dioxide, and Oxygen

Concentrations of carbon monoxide (CO) in the flue gas were determined at the test site by Dräger tube analysis of gas samples collected in Tedlar bags.[3] Field analysis of carbon dioxide (CO_2) and oxygen (O_2) was performed with a Fyrite test kit to determine stack gas molecular weight and excess combustion air.[4]

Organic Compounds and Elemental Emissions

A modified Method 5 procedure was used to collect samples for organic and inorganic species characterization. The modification consisted of the addition of a porous polymer resin trap (XAD-2) after the particulate filter in a standard Method 5 train. The XAD resin, particulate filter and impingers were extracted with methylene chloride and the extract was fractionated on a silica gel column. Each of eight fractions were ultimately analyzed by gas chromatography - mass spectrometry (GC/MS) for organic species including polynuclear organic material (POM).[5]

The extracted particulates were digested using the acid digestion Parr bomb technique.[6,7] After digestion the sample solutions were diluted and submitted for elemental analysis by atomic absorption (AA) and by the AtomComp technique employing an inductively coupled argon plasma (ICAP) excitation source.[8,9]

RESULTS

Test results are presented for each emission species and correlated, when possible, with coal properties. The effects of type of combustion equipment and heating cycle are discussed separately. Analytical results for ash residues are also given.

Emissions

Emission factors for the test conditions used in this program are summarized in Table I for criteria pollutants and POM emissions. Emission factors are, in most cases, for duplicate test runs. As discussed previously, emissions from the furnace were measured independently for the ON and OFF heating cycle segments. These emission rates were combined and are presented as an emission factor for the total heating cycle (20 min ON/40 min OFF). Data for the separate segments are presented later.

Particulates

Prior to this study emission estimates for most coal-fired combustion equipment were based on the ash content of the coal being burned. However, the results of this study indicate that ash content has only a secondary effect on particulate emissions from residential size coal-fired combustion equipment. The primary factors influencing particulate emissions are coal volatile matter content and combustion equipment design.

Table I. Emission Factors for POM and Criteria Pollutants From Coal-Fired Residential Heating Equipment Operated on a 20-min "ON"/40-min "OFF" Heating Cycle.[a]

Heating equipment	Coal designation	Stoker feed rate, lb/hr	Excess air, %	Emission factor, lb/ton						Coal		
				Particulates	SO_x	NO_x	CO	Condensable organics	POM	% ash	% volatiles	% sulfur
Boiler	A-unwashed	21.3	238	5.0	6.0[b]	12.8[b]	0.26	2.0	0.26	10.9	39.1	0.41
Boiler	A-blend	22.0	123	7.8	11.0[b]	7.0[b]	4.0	2.4	-[c]	7.5	40.5	0.38
Boiler	A-washed	21.6	171	6.8	9.6[b]	7.6[b]	0.30	2.2	0.58	4.3	42.3	0.42
Boiler	B-unwashed	21.1	128	2.8	17.8[b]	2.8[b]	0.08	2.4	-[c]	9.1	37.5	1.5
Boiler	B-Blend	17.2	151	4.8	30[b]	4.6[b]	0.08	3.6	-[c]	7.1	38.8	0.97
Boiler	B-washed	15.4	160	5.8	14[b]	6.4	0.08	5.0	-[c]	5.0	38.7	0.58
Furnace	B-wash	15.6	129	44	12.6	13.6	24	-[c]	0.070	5.0	38.7	0.58
Furnace	B-blend	15.0	-[c]	20	30	13.2	8.8	9.4	-[c]	7.1	38.8	0.97
Furnace	B-unwashed	15.8	117	26	28	7.8	26	5.2	-[c]	9.1	37.5	1.5
Furnace	B-unwashed	15.8	182	34	30	6.0	26	-[c]	0.036	9.1	37.5	1.5
Furnace	C-as mined	22.0	207	4.0	-[c]	-[c]	-[c]	3.6	-[c]	3.3	34.7	0.47

[a] Most emission factors represent the average of duplicate sampling runs.

[b] Data for ON segment of heating cycle only.

[c] No data obtained due to program limitations.

Particulate emissions were found to vary inversely with coal ash content, contrary to previous estimating procedures, but did increase proportionally with increasing coal volatile content. Analysis performed on particulate samples revealed that the particulate carbon content was approximately 80%.

Extraction with methylene chloride did not change the carbon content. Consequently, particulate matter is either fragments of unburned coal, nonextractable tars and resins, or carbon black-like material.

Condensable Organics

Condensable organic emissions, collected in the back half of the Method 5 sampling system, ranged from 2.0 lb/ton to 9.4 lb/ton. This accounts for 15% to 50% of the total mass collected by this system. In the past this material has either not been measured or reported collectively with the front half mass as total particulates. The material collected was an amber colored residue resembling coal tars or resins.

Sulfur Oxides

Sulfur oxide emission factors ranged from 6.0 lb/ton to 30 lb/ton. In terms of coal sulfur content, emission factors ranged from 15 S lb/ton to 30 S lb/ton with an average of 22 S lb/ton. (S is the sulfur content of the coal in weight percent.) The relationship of SOx emissions to coal sulfur content is shown in Figure 1. Up to a coal sulfur content of 1.0%, the relationship is linear, but at 1.5% coal sulfur content the SOx emission factors deviated significantly from linearity. These low SOx values may be influenced by the coal lime content. It has been predicted that the lime (i.e., calcium) in coal can influence SOx emissions by combining with sulfur and reducing the formation of SOx. The 1.5% sulfur coal contained 50% more calcium than the 1% sulfur coal.

Nitrogen Oxides

Emission factors for NOx ranged from 2.8 lb/ton to 14 lb/ton. NOx emissions were determined from three 30-sec. grab samples collected during each test run. Wind gusts caused the barometric damper to open and close during testing, and frequently changed the dilution of the stack gases, resulting in a wide variation of NOx levels. Averaging the results of three grab samples for each test run reduced this variability, but not to the extent that would be expected with continuous sampling. Correlation of NOx emissions to test variables was not observed, and it can be reasonably assumed that these emissions will remain unchanged with changes in major variables.

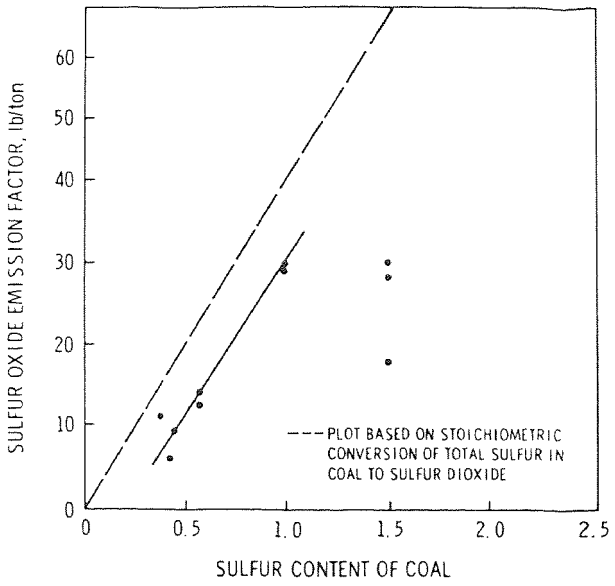

Figure 1. Effect of coal sulfur content on SO_x emissions.

Carbon Monoxide

Carbon monoxide emission factors ranged from less than 0.08 lb/ton to 26 lb/ton. This large variability could not be explained completely by known variables. However, such variability has also been observed in tests conducted by other researchers. Apparently CO is the emission most sensitive to changing combustion conditions and indicates the instability and nonuniform nature of the fuel bed.

Polynuclear Organic Material

Emission factors for polynuclear organic materials ranged from 0.036 lb/ton to 0.58 lb/ton. Combustion of Coal A in the boiler resulted in an average POM emission factor of 0.42 lb/ton, while combustion of Coal B in the warm-air furnace resulted in an average POM emission factor of 0.052 lb/ton. Whether this difference is due to the coal types or combustion equipment cannot be determined from the data. It would be expected that higher volatile content coals would have the potential for emitting greater quantities of POM's.

POM emission factors versus coal volatile content are presented for this study and for one conducted by Battelle[1] in Figure 2.

Points representing MRC data are for one test each, while the Battelle points are averages of four tests each. The Battelle study showed a high variability of POM emission factors for each test condition, indicating the influence of other variables. Figure 3, showing POM emission factors versus excess air, indicates that high excess air reduced POM emissions.

Major Organic Compounds

Organic compounds identified in the flue gas emitted from the boiler are presented in Table II along with their emission factors. Over 45 organic species were identified, 20 of which are POM compounds.

A comparison of the total emission factors for all organic species (6.0 lb/ton) with the emission factor for condensable organic material reported earlier (2.0 lb/ton to 9.4 lb/ton) shows excellent agreement. It is concluded therefore that Table II lists all the major chromatographable organic species emitted from the residential combustion units during this test program.

A special effort was made to detect polychlorinated biphenyl compounds (PCB's) by GC/MS, but none were found in any of the runs. Based on the sensitivity of the instrument, it can be concluded that if such compounds are present, they must have emission factors less than 10^{-5} lb/ton.

Individual Elements

Combustion releases to the atmosphere a portion of the elemental constituents found in coal. Emissions of individual elements in the feed coal in Table III. Values are presented as pounds of element per ton of coal.

Elemental emissions are limited by the amount of elements in the coal. The fractions of elements in the coal emitted to the air are a function of the chemical and physical properties of the elements and the conditions in the combustion equipment. The mass of elements emitted to the atmosphere represent less than 10% of each element in the coal feed except in the cases of those elements typically enriched in the particulates or potentially volatilized during combustion. The exceptions are the large quantities of silicon and zinc that were measured. It is suspected that contamination from glass containers accounts for the silicon, while zinc is probably emitted from erosion of the galvanized exhaust stack.

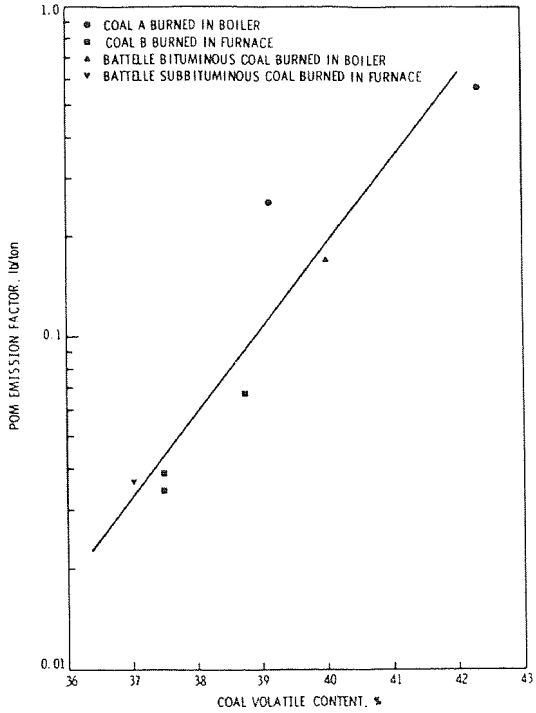

Figure 2. Effect of coal volatile content on POM emissions.

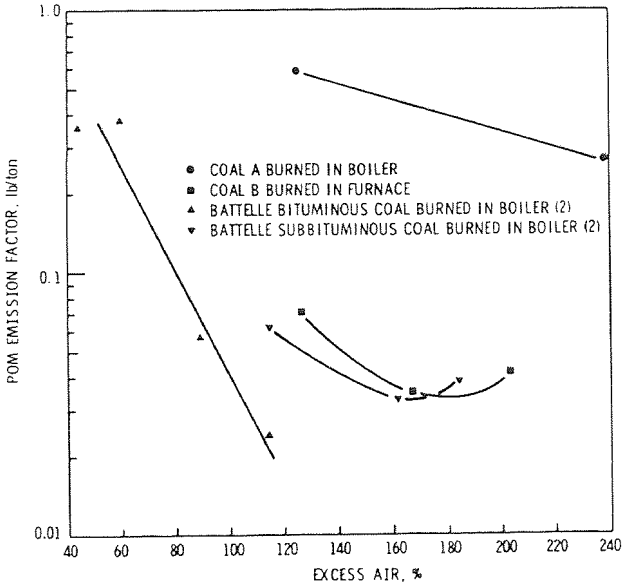

Figure 3. Effect of excess air on POM emissions.

TABLE II. Emission Factors of Major Organic Species From Residential Combustion of Coal (lb/ton)

Identified compound	Coal A
Indane	0.004
Indene	0.050
Methylindenes	0.058
Phenol	0.108
Methyl phenol	0.088
Dimethyl phenol	0.032
Naphthalene	0.56
Methylnaphthalenes	0.44
Dimethylnaphthalenes	0.40
C3-alkyl-naphthalenes	0.176
C4-alkyl-naphthalenes	0.056
C5-alkyl-naphthalenes	0.016
Biphenyl	0.018
Acenaphthalene	0.144
Fluorene	0.096
Methylfluorene	0.064
Phenyl phenol	0.008
Methylbenzofuran	0.04
Fluorenone	0.042
Anthraquinone	0.010
Methyl laurate	0.004
Methyl myristate	0.004
Methyl palmitate	0.012
Methyl stearate	0.006
Di-C3-alkyl-phthalate	0.0008
Di-C4-alkyl-phthalate	0.0012
Di-ethylhexyl-phthalate	0.024
Di-octyl-phthalate	0.30
Aliphatics (C14 to C33)	2.6
POM:	
Dibenzothiophene	0.0006
Anthracene/phenanthrene	0.058
Methylanthracenes/phenanthrenes	0.030
9-Methylanthracene	<0.0004
Dimethylanthracenes/phenanthrenes	0.02
Fluoranthene	0.02
Pyrene	0.018
Methylfluoranthenes/pyrenes	0.008
Benzo(c)phenanthrene	0.0008
Chrysene/benz(a)anthracene	0.014
7,12-Dimethylbenz(a)anthracene[a]	0.30
Benzofluoranthene(s)	0.014
Benzopyrene(s) (and perylene)	0.012
3-Methylcholanthrene[a]	0.006
Indeno(1,2,3-cd)pyrene	0.008
Dibenz(a,h)anthracene[a]	0.012
Dibenzo(c,g)carbazole	<0.0004
Dibenzopyrenes	0.034
Methylchrysenes[a]	0.02
Total	6.0

[a]May include isomers.

TABLE III. Elemental Emission Factors from Coal-Fired Residential Heating Equipment Compared With Coal Elemental Content.
(lb/ton)

	Test Coal B (as received)		Emission factor	
Element	Washed	Unwashed	Unwashed[a]	Washed[a]
Aluminum	9.8	12	0.17	0.18
Antimony	0.028	0.28	0.054	<0.010[b]
Arsenic	0.006	0.01	0.004	0.002
Barium	0.26	0.32	0.014	0.014
Boron	5.0	1.5	0.016	0.050
Cadmium	<0.014[b]	<0.014[b]	0.0002	<0.001[b]
Calcium	8.4	13	0.28	0.34
Chromium	0.086	0.078	0.001	0.006
Cobalt	0.004	0.03	0.001	0.001
Copper	0.066	0.11	0.008	0.004
Iron	6.0	12	0.22	0.19
Lead	<0.14[b]	<0.14[b]	0.002	0.024
Magnesium	3.0	3.4	0.072	0.11
Manganese	0.042	0.096	0.001	0.002
Mercury	<0.00002	<0.00002	0.0006	<0.00002[b]
Molybdenum	<0.056[b]	<0.056[b]	<0.066[b]	<0.014[b]
Nickel	0.34	0.20	0.012	0.068
Phosphorus	0.62	0.70	0.014	0.042
Selenium	<0.072	0.024	<0.006[b]	<0.007[b]
Silicon	0.28	0.24	0.17	0.19
Silver	0.054	0.03	0.12	0.010
Sodium	4.0	0.42	0.001	0.024
Strontium	0.13	0.34	0.004	0.002
Tin	<0.11[b]	<0.11[b]	0.014	0.14
Titanium	0.60	0.94	0.010	0.016
Vanadium	0.11	0.13	0.004	0.006
Zinc	<0.0008[b]	<0.006[c]	0.04	0.016

[a]Average of duplicate runs.

[b]Less than the detection limit.

[c]Less than reagent blank.

Ash Residue

The quantity of ash residue recovered ranged from 220 to 600 pounds per ton of coal burned. Based on the coal ash content the combustion residue is at least 25% nonash components (rocks, unburned coal, etc.). Three samples of residue were analyzed and found to contain from 15% to 56% volatile and combustible material, or 44% to 85% actual ash. This accounts for the high variability of residue recovery and indicates the incomplete nature of the combustion process.

An unambiguous evaluation of the effect of combustion equipment on emissions would require that all other variables be held constant. It was impossible to accomplish this in the present study because of the large number of variables and limited number of tests. However, the effect of many variables appears less significant than the effect of the combustion equipment, and certain observations can be made.

In general, the furnace generated more emissions than the boiler, except in the case of POM's where measurements were not made on identical fuels. Particulate emisions were eight times higher from the furnace than from the boiler. Emissions of SO_x were essentially the same, in support of the hypothesis that variables other than coal sulfur and calcium content exert little if any influence on SO_x emissions.

NO_x emissions were twice as high from the warm-air furnace as from the boiler. The higher values from the furnace indicate that the furnace may have operated at higher temperatures and/or longer residence times. Carbon monoxide emissions from the furnace were almost two orders of magnitude greater than those from the boiler.

Effect of Heating Cycle

Automatic coal-fired heating equipment differs from other forms of automatic heating in that a bed of fuel continues to burn when the demand for heat is satisfied and the combustion-air fan shuts off. Consequently the equipment continues to release pollutants to the atmosphere during the OFF period.

Emission rates for all emission species were higher during the OFF segment. Because POM emissions are products of incomplete combustion of volatiles, this finding was not unexpected.

Emission rates for particulates were about five times higher during the ON segment due to more rapid combustion and higher air velocities through the fuel bed that entrained more particles in the flue gas. Elemental species and NO_x were

emitted at rates 17 and 5 times higher, respectively, during the ON segment.

Emissions data for this study were determined for heating equipment operating at a 20-min ON/40-min OFF heating cycle. Furnaces controlled by thermostats may have ON/OFF cycles of differrent duration. Linear extrapolation of the data obtained in this study to predict emissions for other heating cycles is possible if the emission rates are not strongly dependent on the length of the ON and OFF periods.

CONCLUSIONS

This investigation represents one of the most comprehensive programs undertaken to characterize emissions from coal-fired residential combustion equipment. Nevertheless, test results apply only to the types of combustion equipment and varieties of coal sampled. Extrapolating these results to other test conditions should be done carefully. Further study on different combustion systems and a broader range of coal types would provide valuable information on variations in emissions, and is to be recommended.

Emissions data collected during this study are believed to be typical of what would actually be observed in homes heated with coal. Combustion systems, coal quality and operating conditions were all chosen to fall within, although not encompass, the normal range expected in the residential sector. Test results show that all three parameters influence emission rates, and that there is a wide variation in emission rates even under normal conditions.

In contrast to previous predictions, particulate emissions were not a function of coal ash content but instead correlated with coal free swelling index and volatile content. Whereas in larger combustion systems particles are formed from inorganic ash in the coal, in these residential units particles were formed from the organic material in the coal, and the particulate composition averaged about 80% carbon. This behavior is a consequence of incomplete combustion in the residential units studied.

Because combustion efficiencies for coal-fired residential combustion systems were lower than those for larger units such as utility boilers, emissions of organic species were relatively high. Ash residues contained a high percentage of unburned carbon and, as just mentioned, particulate emissions were primarily carbonaceous materials. Because the organic emissions included a number of carcinogenic polynuclear organic

compounds (e.g., benzopyrene), the trend toward greater residential coal usage may have a significant impact on local air quality. It is suggested that this impact be evaluated in terms of its potential health and environmental effects before a large number of homeowners install coal-fired heating equipment.

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