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# EMISSIONS of POLYNUCLEAR HYDROCARBONS and other POLLUTANTS from HEAT-GENERATION and INCINERATION PROCESSES\*

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Recognizing the growing need for specific emission data from a multitude of air pollution sources, the U. S. Public Health Service, Division of Air Pollution, has undertaken a program of field sampling to obtain measurements of pollutant emission levels from a wide variety of sources.

Under this source-sampling program, emissions to the atmosphere from many chemical, metallurgical, and combustion processes will eventually be evaluated. This paper presents emission data from the first series of tests, for which the sources tested included typical combustion processes involving the burning of conventional fuels (coal, oil, and gas) and of certain commercial and municipal solid wastes. In addition to obtaining over-all emission data from different sizes and types of combustion units, a primary objective of the study was to establish the relative importance of various combustion processes as contributors of benzo(a)pyrene (3,4-benzopyrene) and other polynuclear hydrocarbons with demonstrated or potential carcinogenic properties. The other pollutants measured included particulate matter, carbon monoxide, total gaseous hydrocarbons, oxides of nitrogen, oxides of sulfur, and formaldehyde.

### Sources Tested

Two categories of combustion sources were tested. Those burning conventional fuels were designated as heat-generation processes, and those burning waste materials were classed as incineration processes. Design and operation data for the units studied are given in Tables I and II.

The heat-generation sources tested ranged in size from heavy industrial power plant boilers to residential heaters and employed the following firing methods: (1) pulverized coal, chain grate stokers, spreader stokers, under-feed stokers, and hand-stoked coal burners; (2) steam-atomized, air-atomized, centrifugal-atomized, and vaporized oil burners; and (3) premix gas burners. The incineration sources tested ranged from large municipal incinerators to small commercial incinerators. Open-burning sources were also tested.

### Sampling and Analytical Techniques

Sampling of flue gases was performed while the combustion processes were operated at their normal burning condition. Sampling points were located in the stack or breeching, depending on accessibility, and in the smoke plumes of the open-burning sources.

### Particulate Matter and Polynuclear Hydrocarbons

In collection of samples for polynuclear hydrocarbon analysis, the gas stream was passed through a series of water bubblers kept at 32°F (Fig. 1). These were followed in sequence by (1) a series of freeze-out traps immersed in a dry-ice-alcohol bath held at -98°F, and (2) a high-efficiency glass-fiber filter (MSPA type 1106-BH). This sample-collection technique was designed to minimize loss of the condensed polynuclear hydrocarbons subjected to a continuous flow of gas during extended sampling periods.<sup>1,2</sup> Before the collection sample was subjected to benzene extraction, the total weight of particulate collected was determined by burning the following reaction:  $C_xH_y + (x + \frac{y}{4})O_2 \rightarrow xCO_2 + \frac{y}{2}H_2O$

- (1) Material brushed from the sampling probe.
- (2) Material collected on the sampling-train filter.
- (3) Material filtered from the condensate (and liquids (distilled water and redistilled benzene) used to rinse the train).
- (4) Material recovered from evaporation of the condensate and water rinses.

These samples were collected under isokinetic sampling conditions at rates ranging from four to six scfm,\* and most sources were sampled continuously over periods of one to three hours. The weight of material collected ranged from 0.072 to 30 g. On small stacks, samples were collected at a point of average gas velocity. On larger stacks, the gas stream was traversed, and the sample collection rate was based on the local velocity at each position.

Samples from open-burning sources were collected in MSA type 1106-BH glass-fiber filters. The sampling equipment consisted of a high-volume air sampler (up to 50 cfm) attached to the end of an eight by 10-inch duct, the open end of which was positioned in the smoke plume. Wet burlap wrapped around the duct served to cool the gas stream ahead of the filter. The resulting particulate samples, while indicating the percentage of individual polynuclear hydrocarbons in the particulate emissions, do not indicate total emissions since combustion-gas volume was not measured and sampling was not done under isokinetic conditions.

The initial step in the analytical technique used for determining polynuclear hydrocarbons involved benzene extraction of the particulate matter, the equilibrium, and the subsequent separation of the benzene-

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\* Standard cubic feet per minute at 70°F and 29.9 in. Hg.

soluble fraction of the samples was done by column chromatography, and the analysis was made by ultraviolet-visible spectrophotometry.<sup>3,4</sup> The individual polynuclear compounds that were quantitatively analyzed include benzo(a)pyrene, anthracene, phenanthrene, pyrene, fluoranthene, benz(a)anthracene, benzo(e)pyrene, perylene, benzo(ghi)perylene, anthanthrene, and coronene. As indicated in Table III, these compounds can be placed in two groups corresponding to the relative reliability of the analytical determination for the individual compounds. Because of interferences that occur in the spectrophotometric curves, values for the compounds in Group 2 of Table 3 may be of lower reliability than those in Group 1. Collection efficiency of the sampling train was 99% or better for all of the polynuclear compounds studied.

### Carbon Dioxide, Oxygen, Carbon Monoxide and Total Gaseous Hydrocarbons\*

CO<sub>2</sub>, O<sub>2</sub>, CO and total gaseous hydrocarbon concentrations were measured by collecting a 50- to 100-liter sample of the combustion gases in a Mylar plastic bag and subjecting this sample to individual analyses. Integrated samples were obtained by maintaining a flow of one liter per minute or lower of sample gas to the bag over a one-to three-hour period simultaneous with the polynuclear hydrocarbon sampling. Where different modes of operation yielded more than one flue-gas flow rate in the stack, the sample flow rate to the bag was adjusted proportionately.

\* The symbol "NO<sub>x</sub>" is used to denote oxides of nitrogen determined as equivalent NO<sub>2</sub>.

\* The term "total gaseous hydrocarbons" is used to denote those organic compounds remaining in the gas phase at room temperature in a Mylar plastic bag.

CO<sub>2</sub> was analyzed by Orsat and checked with a nondispersive infrared analyzer; O<sub>2</sub> was measured by Orsat and a paramagnetic type O<sub>2</sub> analyzer; CO was measured by a nondispersive infrared analyzer and gas detector

tubes. Total gaseous hydrocarbon was measured by a flame ionization analyzer.

### Oxides of Nitrogen

NO<sub>x</sub>\* concentrations were measured from two-liter integrated samples analyzed by either the Saltzman<sup>5</sup> or the

Table I—Design and Operational

Source No.	Fuel Used	Firing Method	Type of Unit	Utilization	Design Data	
					Rated Capacity per hr	10 <sup>3</sup> L5 Million Btu*
1	Coal	Pulverized (dry bottom furnaces) → 14A @ 95% eff.	Water-tube boiler	Electric generation	1060	
2				Process heating	200	
3		Chain grate 4.5A	Water-tube boiler	Electric generation	125	
4		Spreader stoker (with reinjector) 5A (often cyclone) → 25A @ 80% eff.		Process heating	70.5	10000 m <sup>3</sup> @ 150. =
5	TOTAL PART.	Underfeed stokers	Fire-tube boiler	Process heating → 3.5A		7.2
6				School heating → 1.4A		3.8
7			Cast-iron sectional boiler	Home heating → 3.1A		0.25
8			Hand-stoked 3.2A ton	Hot-air furnace	Home heating	
9	Oil	Steam-atomized	Water-tube boiler	Process heating	22	23
10					36	30
11		Low-pressure air-atomized	Scotch-marine boiler	Hospital heating		4.2
12		Centrifugal-atomized	Cast-iron sectional boiler	Home heating		0.25
13			Rot-air furnace			0.14
14		Vaporized	Hot-air furnace	Home heating		0.09
15	Gas	Premix burners	Fire-tube boiler	Process heating		7.2 - 225 ehp
16			Scotch-marine boiler	Hospital heating		4.2
17			Double-shell boiler	Home heating		0.18
18			Hot air furnace			0.21
19			Wall space heater			0.625

\* Gross heat input.

Table II—Design and Operational Summary, Incineration and Open-Burning Sources

Source No.	Type of Unit	Rated Capacity, Tons/Day	Design		Fuel Type	Moisture Content, %	Operating Conditions			During Test	
			Dust Collector	Grate Area, Ft <sup>2</sup>			Charging Rate, Tons/Day	Underfire Air, %	Excess Air, %	Gas Temp., °F	Smoke, % Capacity
20	Municipal incinerators Multiple chamber, traveling grate (continuous feed)	250	288	Settling chamber	Residential refuse (14 to 20% non-combustible)	35	260	80	185*	1940*	20
21	Multiple chamber, batch charged, reciprocating stoker grates	50	85.5	Water spray scrubber	Residential and commercial refuse (14 to 20% non-combustible)	25	49	50-60	108*	1470*	
22	Commercial incinerators Single chamber	5.3	13	None	Cardboard, packing crates	20	4.7	0	465*	1300*	0-20
23	Multiple chamber with auxiliary gas burner in primary chamber	3	19.7	None	60% paper 40% wet garbage	50	2.3			1850*	0-50
24	Open Burning				Municipal refuse	20					20-300
25					Automobile tires						20-100
26					Class clippings, leaves, tree branches						20-100
27					Automobile bodies						20-100

h. breaching.  
 i. In furnace.  
 j. In stack.

## Summary—Heat Generation Sources

Dust Collector	Fuel Data as-received basis			Operating Conditions During Test				
	Volatile, %	Ash, %	S, %	Fuel Rate	Gross Btu Input	Steam Rate	Steam Pressure, Psig	Smoke, Opacity, %
				Lbs	Per Hr Million Btu	10 <sup>3</sup> Lb		
Mechanical electrical	31	20.2	2.3	132,000	1560	1120	2000	30-40
Multiple cyclone	36	4.3	0.9	9,420	130	106	397	60
None	44	7.0	3.8	12,400	147	111	450	20-40
Multiple cyclone	37	4.7	0.8	4,290	59.2	49	160	0-20
None	36	4.7	0.7	317	4.4		110	20-40
	19	5	0.8	214	3.0		37	0-20
	38	3.9	1.0	4.8	0.066		9	0-20
	38	2.7	0.5	8	0.115			40-80
None	No. 2 Fuel Oil (28.5° API)	3.2		1,110	21	17.9	250	5
	No. 6 Fuel Oil (13.5° API)	0.7		769	14.4	10.3	125	5
	No. 1 Fuel Oil (43.5° API)			35	0.70		95	0
	No. 2 Fuel Oil (31.5° API)			8.8	0.17			0
	No. 2 Fuel Oil (31.5° API)			4.4	0.085			0
	No. 1 Fuel Oil (43° API)	0.05		1.2	0.025			0
None	Natural gas (94.2% methane 3.6% ethane)			402	9.3		108	0
				42	0.98		96	0-20
				7.9	0.18			0
				7.4	0.17			0
				0.52	0.012			0

phenoldisulfonic acid technique.<sup>6</sup> The Saltzman technique was not satisfactory for analyzing samples from units that burn fuel with significant sulfur content because of serious interference from sulfur dioxide.

### Oxides of Sulfur

Samples for SO<sub>x</sub>† analysis were collected by passing the gas stream through a series of three 500-ml Greenburg-Smith impingers containing a three percent solution of hydrogen peroxide.

A sample collection rate of one cfm was maintained over a period of 20 to 40 min. Analysis was done either gravimetrically by precipitating the sulfate as barium sulfate or by titration of an aliquot with barium chloride with thorin as an indicator.<sup>7</sup>

### Formaldehyde (not aldehydes)

Formaldehyde was determined colorimetrically with the chromotropic acid technique by bubbling a sample of flue gas through a series of midjet impingers containing either chromotropic acid or water.<sup>8</sup> Samples were collected at one liter per minute over a period of 20 to 30 min.

### Results

#### Polynuclear Hydrocarbons

Data on polynuclear hydrocarbon emissions from heat-generation sources are summarized in Table IV. Data on emissions from incineration and open-burning sources are summarized in Tables V and VI.

**Coal-Burning Units.**—The polynuclear hydrocarbon emissions obtained from coal-burning units varied widely, depending on the quality of combustion achieved. Small domestic furnaces used for individual home heating produced the highest emission rates. For example, tests on hand-fired and underfeed stoker-fired domestic units, with essentially no combustion controls, showed emissions ranging from 3800 to 400,000 micrograms of BaP\* per million Btu gross heat input. Intermediate-sized units employing chain-grate stokers and spreader stokers achieved better combustion and produced appreciably less polynuclear compounds (26 to 37 micrograms of BaP per million Btu gross heat input).

† The symbol "SO<sub>x</sub>" is used to denote oxides of sulfur determined as equivalent SO<sub>2</sub>.

\* Benzo(a)pyrene, hereafter referred to as BaP.

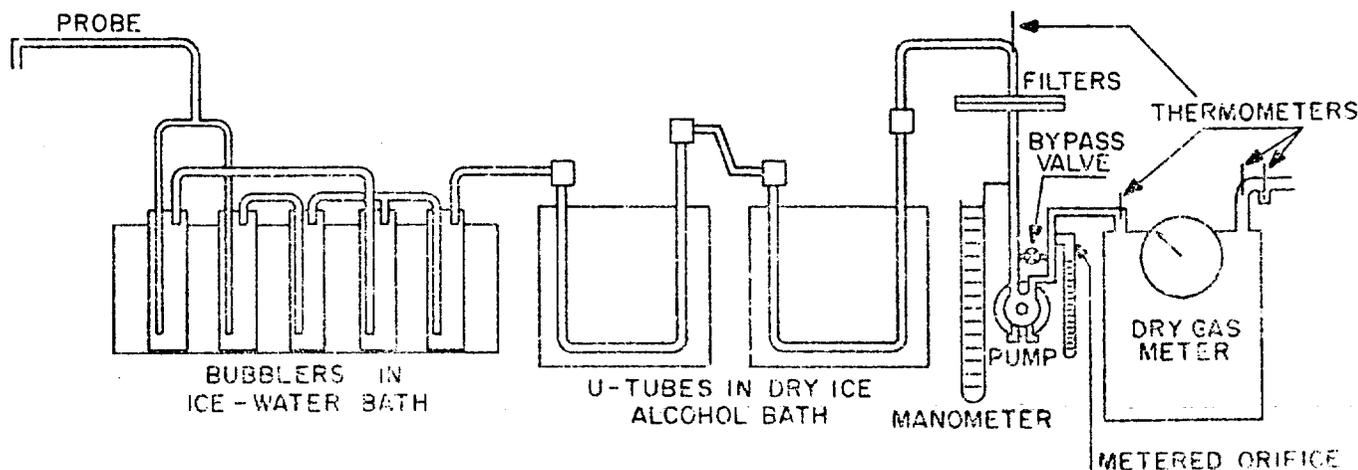


Fig. 1. Polynuclear hydrocarbon sampling train.

Table IV—Polynuclear Hydrocarbon Emission Summary—Heat Generation Sources

Source Fuel No.	Firing Method	Group 1										Group 2	
		Benzo(a)pyrene µgm per 1000 M3 <sup>b</sup>	Pyrene	Benzo(e)-pyrene	Perylene	Benzo(g,h,i)-pyrene	Anthracene	Coronene	Anthracene	Phenanthrene	Fluoranthene	Benzo(a)anthracene	
1	Coal	42	150	92	370	180	19	370	180	19	19		
2	Pulverized	0.22	240	92	370	180	19	370	180	19	19		
3	Chain grate stoker	0.43	32	92	370	180	19	370	180	19	19		
4	Spread stoker	0.44	37	130	390	680	37	390	680	37	37		
5	Underfeed stokers	0.35	26	350	590	360	26	590	360	26	26		
6		140	10,000	7,900	1,600	4,500	290	330	38,000	3,200	3900		
7		61	120	230	1,700	580	1,200	1,000	1,000	47,000	550		
8	Hand-stoked	3,400	3,800	5,400	60,000	300,000	90,000	30,000	1,000,000	1,000,000	1,000,000		
9	Steam-atomized	<38	<0.3	49	<20	300	300	2,100	1,800	56	37		
10	Low-pressure air-atomized	40	0.89	300	47	6,100	300	2,100	3,500	270	37		
11	Centrifugal-atomized	<26	<0.9	1,800	<40	1,800	200	5,300	8,900	5,000			
12	Vaporized	<27	<1	15	<60	15	14	14	15,000	76			
13		<34	<2	1,200	<100	1,200	18	18	77	15,000			
14		<29	<0.4	160	<20	160	14	14	100	100			
15	Premix burners	350	4.6	200	18,000	490	490	5,300	2,900	2,900			
16		<23	<0.5	170	<20	170	170	170	320	320			
17		<30	<0.6	120	<20	120	120	120	110	110			

\* "Less than" values for benzo(a)pyrene were calculated for those samples having concentrations below the limit of quantitative determination (approximately 0.6 microgram per sample). Similar calculations were not included for the other polynuclear hydrocarbons (indicated by blanks in the table).

<sup>b</sup> Micrograms per 1000 cubic meters of flue gas at standard conditions (70°F, 1 atmosphere).

Table V—Polynuclear Hydrocarbon Emission Summary—Incineration Sources\*

Source No.	Type of Unit	Group 1										Group 2	
		Benzo(a)pyrene	Pyrene	Benzo(e)-pyrene	Perylene	Benzo(g,h,i)-pyrene	Anthracene	Coronene	Anthracene	Phenanthrene	Fluoranthene	Benzo(a)anthracene	
20	Municipal 250-Ton/Day Multiple chamber	19	0.575	8.0	0.34	0.24	9.8	0.37	9.8	0.37	0.37		
21	50-Ton/Day Multiple chamber	2,700	6.1	52	12	15	18	4.6	18	4.6	4.6		
	Stack (after scrubber)	17	0.089	2.1	0.58	0.63	3.3	0.15	3.3	0.15	0.15		
22	Commercial 5.3-Ton/Day Single chamber	11,000	53	320	45	3.1	90	6.6	47	140	220		
23	3-Ton/Day Multiple chamber	52,000	260	4200	260	60	870	79	86	59	3900		

\* A blank in the table for a particular compound indicates it was not detected in the sample.

<sup>b</sup> Micrograms per 1000 cubic meters of flue gas at standard conditions (70°F, 1 atmosphere).

**Table VI—Polynuclear Hydrocarbon Content of Particulate Matter Emitted—Incineration and Open Burning Sources\***

Source No.	Type of Unit	Sampling Point	Group 1			Group 2							
			Benzo(a)-pyrene	Pyrene	Benzo(c)-pyrene	Anthracene	Phenanthrene	Fluoranthene	Benzo(a)-anthracene				
20	Municipal incinerators 250-Ton/Day Multiple chamber	Breeching (before settling chamber)	0.016	1.9	0.08	0.06	2.2	0.09					
21	50-Ton/Day Multiple chamber	Breeching (before scrubber) Stack (after scrubber)	3.3 0.15	28 3.6	6.5 0.97	8.2 1.1	9.8 5.5	2.5 0.28					
22	Commercial incinerators 5.3-Ton/Day Single chamber	Stack	58	350	49	3.3	98	7.1	23	51	150	240	5.0
23	3-Ton/Day Multiple chamber	Stack	180	2600	180	36	540	45	130	53	62	2400	210
24	Open Burning Municipal refuse	In smoke plume	11	29	4.5	72	600	53	81	4.7	450	13	560
25	Automobile tires		1100	1300	450		5.4			110		470	25
26	Grass clippings, leaves, branches		35	120	21					4.7		110	
27	Automobile bodies		270	670	120	33	150	12	15	220	100	450	40

\* A blank in the table for a particular compound indicates it was not detected in the sample.

**Table VII—Pollutant Emission Summary—Heat Generation Sources**

Source No.	Fuel Used	Firing Method	Flow, cu ft/hr	Temp. °F	Flue Gas Conditions in Stack			1000 Lb Per Million Btu	Ton of fuel	Total Particulates	Benzene-Soluble Organics	Carbon Monoxide	Hydrocarbons (as Methane)	Oxides of Nitrogen (as NO <sub>2</sub> )	Oxides of Sulfur (as SO <sub>2</sub> )	Formaldehyde			
					H <sub>2</sub> O, %	CO <sub>2</sub> , %	O <sub>2</sub> , %												
1	Coal	Pulverized	415,000	260	5.4	12.3	6.9	0.50	0.56	14.0	0.7	0.004	0.16	0.47	11	88	1.3 × 10 <sup>-4</sup>		
2	Coal	Chain grate	32,300	235	6.2	12.3	6.1	1.90	2.23	61.6	0.3	0.10	0.11	0.30	9.3	265	2.3	62 × 10 <sup>-4</sup>	
3	Coal	Chain grate	35,000	430	0.7	12.1	7.7	0.99	1.31	31.0	0.3	0.51	0.11	0.11	3.2	178	0.9 × 10 <sup>-4</sup>		
4	Coal	Stoker	18,100	405	6.9	10.6	8.5	0.66	0.82	22.6	1.4	<0.1	0.09	0.16	11	143	1.4 × 10 <sup>-4</sup>		
5	Coal	Underfeed	3,340	380	2.1	3.0	17.2	0.68	0.62	17.0	1.1	0.16	0.116	0.30	9.3	265	2.3	62 × 10 <sup>-4</sup>	
6	Coal	Underfeed	3,290	335	2.1	2.5	18.1	0.24	0.25	7.0	3.6	0.14	0.033	0.30	9.3	265	2.3	62 × 10 <sup>-4</sup>	
7	Coal	Hard-soled	43	345	2.2	2.0	17.1	0.52	0.44	12	1.2	0.12	0.12	0.30	9.3	178	0.9 × 10 <sup>-4</sup>		
8	Coal	Hard-soled	78	220	2.1	2.8	17.7	1.80	1.26	37	1.7	0.12	0.12	0.30	9.3	178	0.9 × 10 <sup>-4</sup>		
9	oil	Steam-atomized	5,200	530	8.5	9.0	8.2	0.32	0.305	11.7	1.0	0.055	0.004	0.17	12	188	1.3	48 × 10 <sup>-4</sup>	
10	oil	Low-pressure	10,000	340	5.4	8.8	9.0	0.049	0.055	2.0	7	0.055	0.004	0.17	12	188	1.3	48 × 10 <sup>-4</sup>	
11	oil	Low-pressure	185	230	7.4	8.8	9.0	0.049	0.055	2.0	7	0.055	0.004	0.17	12	188	1.3	48 × 10 <sup>-4</sup>	
12	oil	Atomized	145	170	3.0	2.0	10.0	0.041	0.040	1.8	6.3	0.038	0.004	0.17	12	188	1.3	48 × 10 <sup>-4</sup>	
13	oil	Atomized	115	175	2.6	1.8	18.3	0.070	0.080	3.1	11	0.075	0.021	0.82	2.3	35	0.40	18 × 10 <sup>-4</sup>	
14	oil	Vaporized	49	185	1.9	1.2	19.3	0.067	0.071	2.8	10	0.25	0.030	1.2	4	4	0.08	3 × 10 <sup>-4</sup>	
15	Gas (Municipal)	Premix burner	3,040	380	8.4	3.0	14.3	0.026	0.024	1.0	11	0.013	0.063	0.14	6.4	0.89 × 10 <sup>-4</sup>	0.89 × 10 <sup>-4</sup>	41 × 10 <sup>-4</sup>	
16	Gas (Municipal)	Premix burner	395	210	11.4	5.9	10.0	0.030	0.032	1.5	8.0	0.03	0.052	0.16	7.3	0	0	0	100 × 10 <sup>-4</sup>
17	Gas (Municipal)	Premix burner	92	170	4.8	2.4	16.9	0.010	0.008	0.3	3.3	0.02	0.02	0.08	4.1	0	0	0	110 × 10 <sup>-4</sup>
18	Gas (Municipal)	Premix burner	82	140	4.1	2.2	17.3	0.011	0.007	0.3	2.3	0.025	0.02	0.08	2.8	0	0	0	53 × 10 <sup>-4</sup>
19	Gas (Municipal)	Premix burner	11	205	4.4	2.0	17.5	0.027	0.026	1.2	1.9	0.030	0.016	0.74	0.06	2.8	0.89 × 10 <sup>-4</sup>	0.89 × 10 <sup>-4</sup>	41 × 10 <sup>-4</sup>

\* Blank in the table indicate that no test was made.  
 † Pounds of particulate per 1000 pounds of dry flue gas adjusted to 50% excess air.  
 ‡ Average area of the fuel oils are given in Table I, the density of natural gas = 0.043 lb per cu ft (60°F, 1 atm).

*\* Probably includes methane*  
*Not functioning properly*

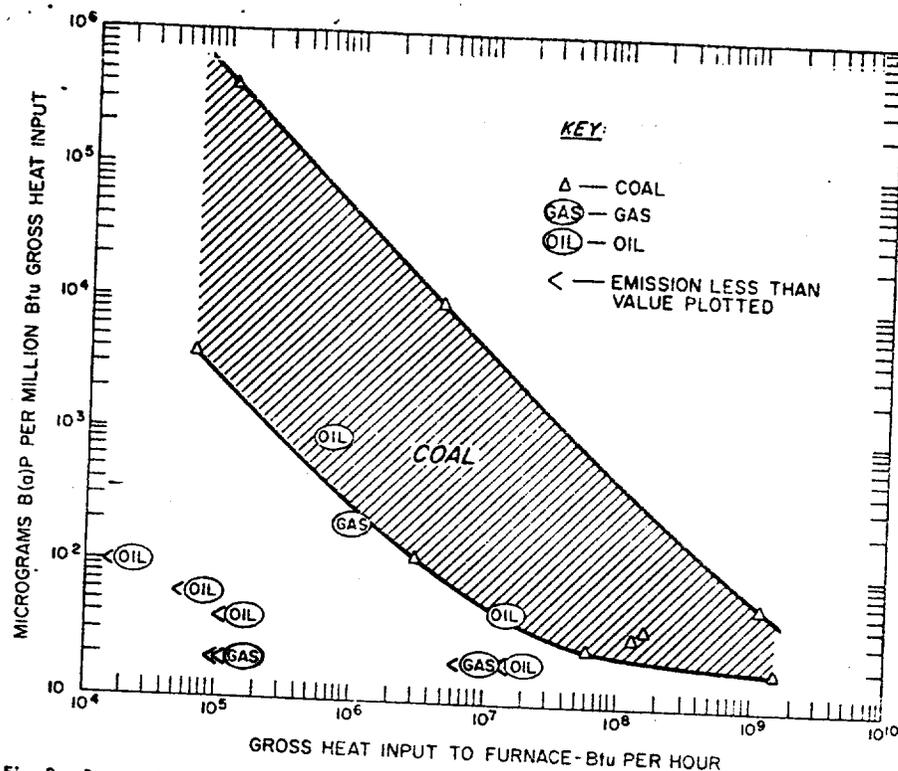


Fig. 2. Range of benzo (a) pyrene emissions from coal, oil, and natural gas heat-generation processes.

The large, fully instrumented, pulverized-coal-fired steam power plant ( $1.06 \times 10^6$  of steam/hr) showed the lowest emission rate (19 micrograms of BaP per million Btu gross heat input).

In Fig. 2, BaP emission rates have been plotted against size of unit for the coal-, oil-, and gas-fired units studied. Because of the many variables affecting the formation of this compound, a wide range of emissions can be expected from a given size of unit, and the results obtained do not lend themselves to a straight-line relationship. In units operating with relatively good combustion conditions, BaP emissions would be expected to fall near the lower boundary of the range, while in less efficient combustion processes emission rates would be expected to fall near the upper boundary. Although it is constructed with a limited amount of data from oil- and gas-burning sources, the plot in Fig. 2 serves to emphasize that higher emissions are to be expected from coal-burning units than from gas- or oil-fired units of comparable sizes.

The lower polynuclear hydrocarbon emission rates from the larger coal-fired units can be attributed to the more efficient combustion of fuel attainable with closely regulated air-fuel ratios, uniformly high combustion-chamber temperatures, and relatively long retention times in the high-temperature zone.

Among the seven polynuclear hydrocarbons for which the analytical technique was most reliable (Table III), pyrene and BaP were the only com-

pounds detected in all coal-fired units studied (Table IV). In each case, the ratio of pyrene to BaP was greater than one, varying from 1.5 to 23. Ratios of BaP to benzo(ghi)perylene ranged from 1.3 to 6.5, and ratios of BaP to coronene ranged from 1.0 to 30. These results tend to confirm the data reported by Sawicki<sup>9</sup> regarding the ratios of these compounds in particulate samples collected from the atmosphere. Sawicki states that particulate pollution from coal-burning sources (as opposed to auto exhaust) might be characterized by ratios of BaP to benzo(ghi)perylene greater than 0.6 and ratios of BaP to coronene greater than one. He further suggests that the ratio of pyrene to BaP might be an indication of the "age" of the particulate pollution, since pyrene is less stable in the atmosphere than is BaP and has a greater tendency to be lost by volatilization. This interpretation is substantiated by the finding that ratios of pyrene to BaP in particulate matter collected from the atmosphere in cities throughout the United States have been reported<sup>10</sup> to be less than one in most cases.

A comparison of polynuclear hydrocarbon emissions with other products of incomplete combustion indicates that polynuclear emission rates are generally high when carbon monoxide and total gaseous hydrocarbons are high. The relationship between carbon monoxide and polynuclear hydrocarbons was not consistent enough to show a good correlation. Figure 3 shows fair correlation between total gaseous hydrocarbons

and the compounds pyrene and BaP.

**Oil-Burning Units.**—Polynuclear hydrocarbon emissions rates for oil-burning sources were generally much lower than from coal-burning sources of equivalent size (see Table IV). Detectable BaP concentrations were found in only two of the six oil-burning units tested, but pyrene was present in all sources. Ratios of pyrene to BaP for the two BaP-emitting units averaged 6.6. Ratios of BaP to benzo(ghi)perylene were >2 and 3.0, and ratios of BaP to coronene ranged from 0.4 to >5. The highest BaP emission rate from oil burning occurred in a unit employing an air-atomized burner.

Low BaP emissions from liquid-petroleum-fired units have also been reported by Howe.<sup>11</sup> In a study of household-sized units (12,000 to 250,000 Btu per hour) and small industrial-sized units (6000 lb of steam per hour), no BaP was detected in the samples collected. Two of the small household-sized units produced smoke during testing.

**Gas-Burning Units.**—Polynuclear hydrocarbon emission rates from gas-fired units tested were generally lower than from coal-fired units and were of about the same magnitude as emissions from oil-fired units. For three of the four gas-fired sources the only compound of consequence was pyrene (Table IV), which occurred at rather consistent levels in all but one source. One gas-fired source (No. 16) was more productive of polynuclear hydrocarbons than the others, apparently because of improper adjustment of the air-fuel ratio as evidenced by high concentrations of CO and total gaseous hydrocarbons (See Table VII). The ratio of pyrene to BaP for this source was 90. The ratios of BaP to benzo(ghi)perylene and BaP to coronene were 0.1 and 0.04, respectively; these values contrast with the higher ratios obtained from coal-fired units and are similar to the lower ratios for auto exhaust.<sup>9</sup>

#### Incinerators and Open-Burning Sources.

**Commercial and Municipal Incinerators.**—Results of the polynuclear hydrocarbon emissions produced by municipal and commercial incinerators are summarized in Tables V and VI. BaP and benzo(e)pyrene, both of which have demonstrated carcinogenic qualities, were detected in the flue gases from every incinerator studied. The concentrations of pyrene from each unit were higher than those of any of the other polynuclear hydrocarbon compounds detected, and ratios of pyrene to BaP ranged from 6.0 to 120. Following the pattern of the coal-fired sources, the small-sized commercial incinerators with relatively poor combustion produced larger emission rates for polynuclear hydrocarbons than the

Table III—Properties of Polynuclear Hydrocarbons

Compound	Empirical Formula	Structure	Molecular Weight	Melting Point, °C	Boiling Point, °C	Biological Activity*
<b>Group 1</b>						
Pyrene	C <sub>16</sub> H <sub>10</sub>		202	150	>360	...
Benzo(a)pyrene	C <sub>20</sub> H <sub>12</sub>		252	178	≈500	+++
Benzo(e)pyrene	C <sub>20</sub> H <sub>12</sub>		252	178	≈500	+
Perylene	C <sub>20</sub> H <sub>12</sub>		252	275	≈500	...
Benzo(ghi)perylene	C <sub>22</sub> H <sub>12</sub>		276	273		...
Anthanthrene	C <sub>22</sub> H <sub>12</sub>		276	257		...
Coronene	C <sub>24</sub> H <sub>12</sub>		300	435	≈600	...
<b>Group 2</b>						
Anthracene	C <sub>14</sub> H <sub>10</sub>		178	217	340	Not reported
Phenanthrene	C <sub>14</sub> H <sub>10</sub>		178	101	340	Not reported
Fluoranthene	C <sub>16</sub> H <sub>10</sub>		202	110	393	...
Benz(a)anthracene	C <sub>18</sub> H <sub>12</sub>		228	160	Sublimes	+

\* Relative activity of mouse epidermis: Code +++ active, ++ moderate, + weak, ... inactive. Symposium on the Analysis of Carcinogenic Air Pollutants. Hoffman, D., and Wynder, E. L. Analytical and Biological Studies on Gasoline Engine Exhaust, National Cancer Institute Monograph No. 9, August, 1962.

intermediate- and large-sized municipal units. The largest municipal unit studied (Source No. 20), operating at relatively constant temperatures in excess of 1600°F and with longer gas retention times in the high-temperature zone, was the least productive of these compounds in terms of emissions per pound of refuse charged.

A water-spray scrubber in the flue-gas exhaust duct for control of fly ash from the smaller municipal unit (Source No. 21) proved highly effective in reducing the level of polynuclear hydrocarbons emitted to the atmosphere. The emission rate of BaP, for example, was reduced 98% by use of the scrubber.

**Open-Burning Sources**—Refuse burning rates and combustion gas flow rates were not obtained from the open-burning sources tested because of the difficulty in making these measurements on an unenclosed fire. Hence, the only

comparisons possible with regard to polynuclear hydrocarbons were on the basis of the amount of such compounds in the particulate matter collected. Table VI summarizes the results obtained from the open-burning sources. The comparatively high levels of all polynuclear hydrocarbon compounds detected in the particulate material collected from open burning of automobile tires and automobile bodies is consistent with the general pattern that has developed for other combustion processes; i.e., the inadequate combustion conditions typical of uncontrolled burning processes consistently produce the highest levels of these compounds.

Particulate emission rates from open burning have been estimated at 22 pounds per ton of combustible refuse burned.<sup>12</sup> This estimate would appear to be a very conservative figure for the particulate material that might be

produced from open-burning sources. The particulate emission rates reported in Table IX and the polynuclear hydrocarbon emission rates reported in Table VI indicate that the amount of polynuclear hydrocarbons per pound of refuse burned are greater from open burning and commercial incineration than from municipal incineration. Very high emissions are indicated from the open burning of automobile bodies and tires.

#### Particulate Matter

**Coal-Burning Units**—Particulate emissions from the coal-burning units are summarized in Table VII. Generalizations concerning these results are not appropriate, since the particulate emission for each individual coal-burning unit is affected by a combination of four highly variable factors:

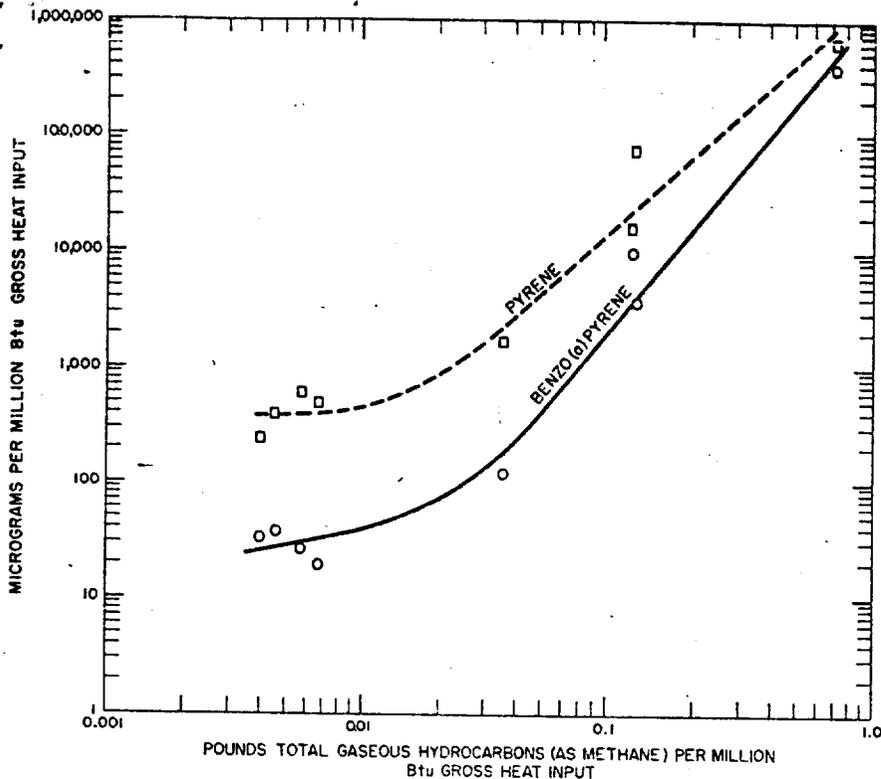


Fig. 3. Relationship between total gaseous hydrocarbon emissions and emissions of benzo (a) pyrene and pyrene from coal-fired heat-generation sources

- (1) Type of firing equipment
- (2) Amount and type of ash in the coal
- (3) Quality of combustion
- (4) Type and suitability of fly-ash-control equipment

In Fig. 4 the particulate emissions for different types of coal-burning units are expressed as a percentage of the total ash content of the coal. The quality of combustion in the larger units was generally better than in the smaller units. Particulate emissions from the large units were primarily of the non-combustible type and amounted to less than 25% of the ash in the coal. Source No. 2, which was a pulverized-coal-fired unit, emitted particulate equivalent to 72% of the ash content in the coal because the mechanical fly-ash collector on this unit was not functioning properly. By comparison, Source No. 1, which is a pulverized-coal-fired unit equipped with properly operating mechanical and electrical fly-ash collectors, emitted particulate equivalent to less than four percent of the ash content in the coal. Although spreader stokers equipped with reinjectors are capable of producing unusually high particulate emissions, Source No. 4, which is equipped with a multicyclone dust collector, emitted particulate equivalent to only 24% of the ash content of the coal.

The emission values shown in Fig. 4, include both combustible and noncombustible portions of the particulate matter emitted. The 69% emission

rate produced by the hand-fired unit (Source No. 8), can be explained in terms of the poor quality combustion typical of these small units, which emit large amounts of unburned carbon and tarry organics. The benzene-soluble portion of the particulate (Table VII), which is a measure of the organic content, ranged from 0.3 to 17%. In general, it is higher for the smaller units; the hand-stoked domestic hot-air furnace (Source No. 8) yielded the highest percentage. By comparison, particulate samples collected from the atmosphere have an average benzene-soluble content of 8.4% for urban areas throughout the United States and 5.6% for nonurban areas.<sup>13</sup>

**Oil-Burning Units.**—Particulate emissions from the oil-fired units, summarized in Table VII, are generally lower than emissions from coal-fired units. The factors of fundamental importance that affect emissions from oil-fired units are:

- (1) Type of firing equipment
- (2) Quality of combustion
- (3) Buildup and/or erosion of boiler deposits
- (4) SO<sub>2</sub> emission

The ash content of the oils burned in this study was too low to be a major factor in contributing to particulate emissions. The highest ash content encountered was 0.025% for the residual fuel oil fired in Source No. 10. The ash content of the distillate fuels was

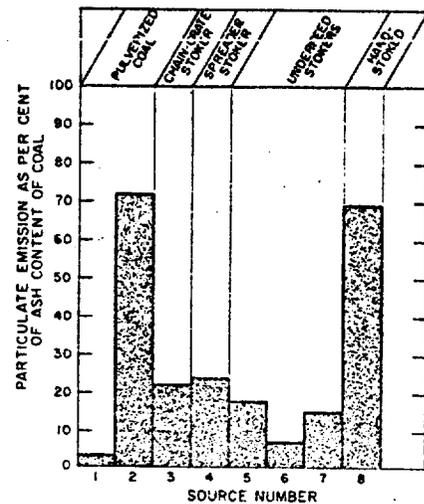


Fig. 4. Ratio of particulate emission to total ash content of coal.

well below this, amounting to only a trace in most cases. Additional factors that affect emissions from oil-fired units include the deposition of particulate material in the boiler as well as the erosion of boiler deposits.<sup>14</sup>

Particulate emissions were three to six times higher on the basis of pounds per million Btu input in the two largest oil-fired boilers tested (23 and 30 million Btu gross heat input per hour) than in the smaller units (0.09 to 4.2 million Btu gross heat input). Both of the large units employed steam atomization and were operated under conditions that produced good combustion. Typical of SO<sub>2</sub> sources, plumes from both units had a bluish-white appearance with about five percent obscuration. A possible explanation for the comparatively high emissions from these furnaces can be made on the basis of the sulfur content of the fuels (Table I). Both units had relatively high oxides of sulfur emissions (Table VII), and SO<sub>2</sub> could account for a good portion of the particulate weight. This possibility was not substantiated, however, since SO<sub>2</sub> concentrations were not measured. The total oxides of sulfur measurements did not differentiate between SO<sub>2</sub> and SO<sub>3</sub>.

The benzene-soluble content of particulates from oil-fired sources was appreciably higher than that from coal-fired sources, ranging from 9.4 to 60% for the small units and from 1.0 to 2.7% for the large units.

**Gas-Burning Units.**—Particulate emissions from gas-fired units are summarized in Table VII. These emissions are lower than from either coal- or oil-fired units. Quality of combustion is the key factor affecting the particulate emissions from the burning of natural gas. Source No. 16, operated with an improper air-fuel ratio, produced the highest particulate emissions.

As was the case with oil firing, the organic portion of the particulate was

Table VIII—Pollutant Emission Summary—Incineration Sources\*

Source No.	Type of Unit	Sampling Point	Flue-Gas Conditions			Total Particulates		Carbon Monoxide	Hydrocarbons (as methane)	Oxides of Nitrogen (as NO <sub>x</sub> )	Formaldehyde
			Flow scfm	Temp. °F	H <sub>2</sub> O %	CO <sub>2</sub> %	O <sub>2</sub> %				
20	Municipal 250-Ton/Day Multiple chamber	Breaching (before settling chamber)	52,000	1270	8.8	6.2	13.7	2.1	0.67	2.5	0.0014
		Breaching (before scrubber)	5,400	1470	9.1	8.7	11.0	1.0	4	<0.08	0
21	Multiple chamber	Stack (after scrubber)	12,700	420	12.4	3.7	16.7	0.34	<2	2.8	0
22	Commercial 5.3-Ton/Day Single chamber	Stack	1,075	1030	9.5	3.5	17.2	0.66	4.3	1.6	0.016
		Stack	580	670	9.3	3.3	16.4	0.96	25	3.6	

\* Blanks in the table indicate that no test was made. <sup>b</sup> Pounds particulate per 1000 pounds dry flue gas adjusted to 50% excess air. <sup>c</sup> Particulate loading in breaching adjusted to indicate emission from stack. <sup>d</sup> Measured by hexane-sensitized nondispersive infrared analyzer; reported as methane. <sup>e</sup> Benzene-soluble?

relatively high compared with that obtained from coal firing, because of the smaller quantities of noncombustible particulates produced by gas and oil firing.

*Incinerators and Open-Burning Sources*

*Commercial and Municipal Incinerators.*—The particulate emissions from incinerators tested are summarized in Table VIII.

The amount of particulate matter discharged from incinerators is affected by a combination of factors:

- (1) Incinerator design
- (2) Type of refuse
- (3) Refuse-charging rate
- (4) Quality of combustion
- (5) Ratio of underfire air to overfire air
- (6) Control equipment

The two municipal incinerators tested were well designed, and both achieved good combustion when properly operated. The problem of particulate emission from these units, therefore, was essentially one of noncombustible fly-ash emission. The high gas temperatures and velocities inherent in these units are conducive to formation and discharge of fly ash. These emissions can generally be reduced by proper adjustment of the underfire-overfire air distribution and/or by use of fly-ash control equipment. The water spray scrubber on the 50-ton-per-day unit, for example, reduced the particulate emission by 67%.

The situation with regard to the two smaller commercial units tested was quite different. On these incinerators the quality of combustion was the controlling factor. The smoke plumes from both units ranged from grey to black, an indication of unburned carbon. Furthermore, these flue gases were highly odorous, an indication of emissions of unburned organic compounds.

Although the three-ton-per-day incinerator (Source No. 23) was equipped with an auxiliary gas burner in the primary combustion chamber, it still produced higher emissions than did the 5.3-ton-per-day single-chamber unit (Source No. 22). This was caused by (1) burning of refuse with high moisture content (50% by weight vs 20% for Source 22), and (2) adjustment of the barometric damper to allow a too-rapid influx of ambient air, which cooled the combustion gases prematurely.

The organic content of particulate emissions from incinerators, as indicated by the benzene solubility, amounted to 0.3% for the two large units and 1.3 to 4.4% for the small units.

*Open-Burning Sources.*—Particulate matter collected from the open-burning sources contained a higher percentage of organic material than that produced by

any of the enclosed-burning sources. The benzene-soluble content was 65% for municipal refuse and 73% for grass and hedge clippings. The collected particulate was yellow-brown in the first case and grey-brown in the second, an indication that only small amounts of carbon soot were present. Particulate produced by the open burning of rubber tires was 15% benzene-soluble, and the particulate produced by the burning of automobile bodies was 48% benzene-soluble. For both of these sources the black color of the collected particulate indicated a large percentage of carbon soot.

*Carbon Monoxide and Total Gaseous Hydrocarbons*

*Coal-, Oil-, and Gas-Burning Units.*—The quantity of CO or total gaseous hydrocarbons emitted from a combustion process is dependent on the quality of combustion. Many of the factors that affect the quality of combustion are better controlled on larger boiler installations, which yield lower emissions per unit of fuel as shown in Table VII. Emissions from the oil-fired units tested are generally lower than from coal-fired units, and emissions from the gas-fired units were still lower except for those from Source No. 16, which had a poorly adjusted burner.

The degree to which emissions of total gaseous hydrocarbons and carbon monoxide follow the same general trend is shown in Fig. 5. Emissions of total gaseous hydrocarbons are plotted against carbon monoxide emissions; the least-squares line drawn through the points has a correlation coefficient of 0.71 (significant at the 1.0% level). A better correlation between CO and total gaseous hydrocarbons was not expected in view of the variety of types of combustion processes involved. Hydrocarbons are more susceptible to oxidation and decomposition at high temperatures than is carbon monoxide. Accordingly, the hydrocarbon emission rates are lower than those for carbon monoxide, and in general the ratio of the total gaseous hydrocarbon emission to the CO emission is lower for the larger units.

*Incinerators.*—The same relationship between combustion quality and emissions of CO and total gaseous hydrocarbons holds true for incinerators; the small commercial-sized units had the highest emission rates (Table VIII).

*Oxides of Nitrogen*

*Coal-, Oil-, and Gas-Burning Units.*—NO<sub>x</sub> emissions for the least-generation sources tested are shown in Table VII. In Fig. 6 the rate of NO<sub>x</sub> emission for each unit is plotted against gross heat input to the furnace. The least-squares line drawn through the points has a slope of 1.14 and a cor-

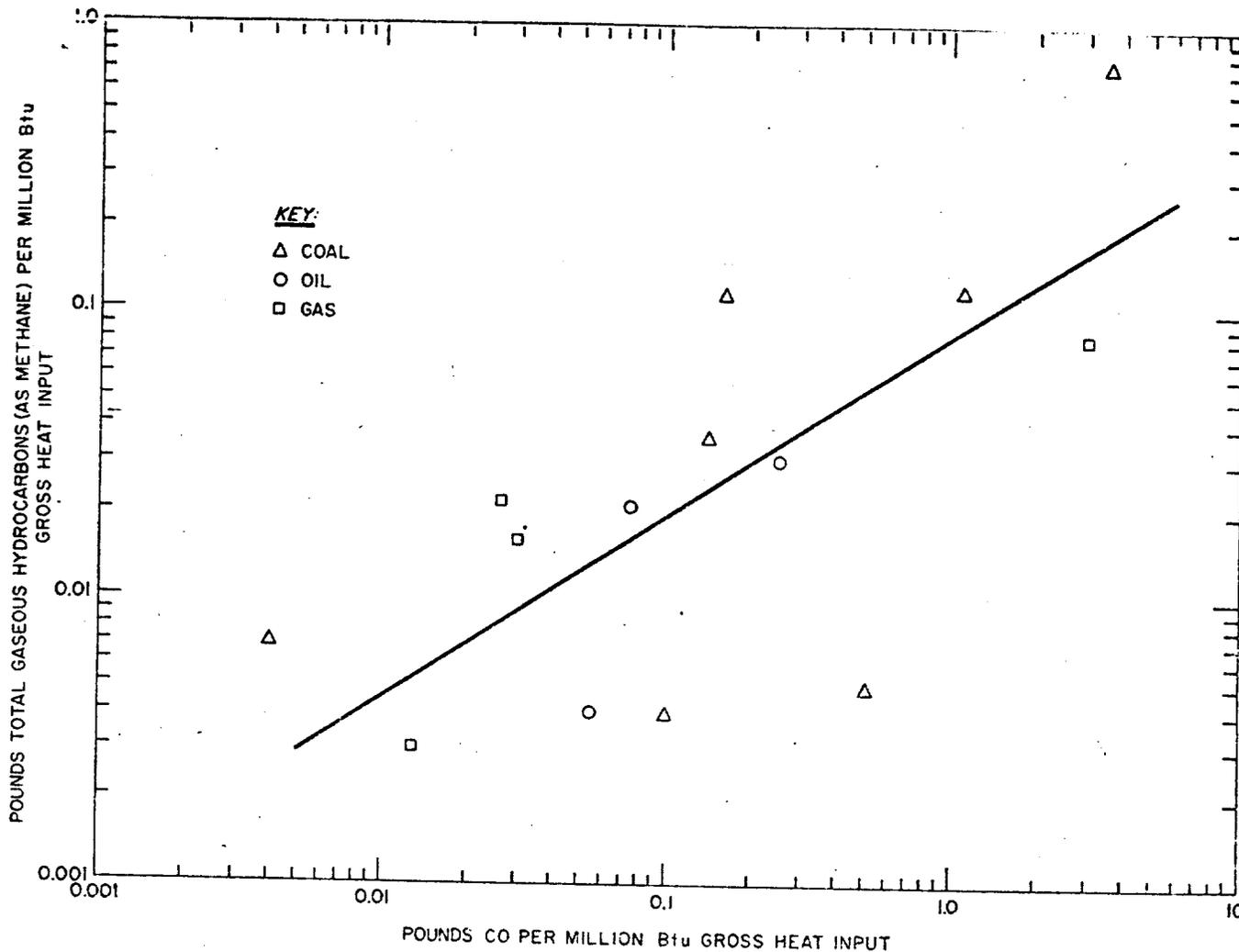


Fig. 5. Relationship between emissions of carbon monoxide and total gaseous hydrocarbons—heat generation sources.

relation coefficient of 0.98 (significant at the 0.1% level). Plotted on the same graph are curves reported by other investigators for oil- and gas-fired units.<sup>15</sup> Each of these curves indicates that the rate of  $\text{NO}_x$  emission is a power function of the gross heat input with an exponent greater than 1.0; hence the larger units are more productive of  $\text{NO}_x$  on a pound-per-Btu basis. This effect is to be expected since higher temperatures in these larger units are a major factor in producing increased  $\text{NO}_x$  emissions. Lower-than-average emissions for the majority of the gas-fired units (Fig. 6) are also in agreement with the results reported by the other investigators.

**Incinerators.**— $\text{NO}_x$  emissions from the incinerators tested are summarized in Table VIII. Higher emissions per unit of fuel were produced by the large municipal incinerators. This trend is analogous to that found for the heat-generation sources.

A more detailed evaluation of  $\text{NO}_x$  emissions from the municipal incinerators was previously reported in the literature.<sup>16</sup>

#### Oxides of Sulfur

##### Coal-, Oil-, and Gas-Burning Units.

$\text{SO}_x$  emission data for the heat-generation sources are presented in Table VII. Flue gases from all of the coal- and oil-burning units tested contained sulfur oxides. No  $\text{SO}_x$  was found in the effluents from the natural-gas-burning units tested.

In Fig. 7,  $\text{SO}_x$  emissions are plotted against sulfur content of the fuel. Although the data obtained for sulfur content of the fuels are not precise values, the graph indicates that roughly 80 to 100% of the sulfur in the fuels burned was converted to sulfur oxides.

#### Formaldehyde

**Coal-, Oil-, and Gas-Burning Units.**—Formaldehyde emissions from heat-generation sources are summarized in Table VII. Since formaldehyde is an oxygenated hydrocarbon, emissions of this compound might be expected to correlate well with total gaseous hydrocarbon emissions. Formaldehyde, however, is more susceptible to oxidation and decomposition at the high temperatures encountered in combustion than are some of the lower hydrocarbons, such as methane, and thus the levels of formaldehyde emission were consistently low, ranging from  $1/50$  to  $1/500$  of the more widely varying emissions of total gaseous hydrocarbons.

For each fuel the smaller units tend toward higher emissions; the highest emission (0.66 ppm) occurred in the flue gas of the small gas-fired space heater (No. 19).

**Incinerators.**—Formaldehyde emissions from the incinerators are reported in Table VIII. The 50-ton-per-day incinerator, although it produced no formaldehyde at the conditions reported here, did produce 0.021 pound per ton of refuse when operated with a higher amount of excess air. Operation at this latter condition resulted in furnace-gas temperatures 400°F lower than those that occurred in the first test. This effect corresponds to results obtained previously with an experimental incinerator, which demonstrated that formaldehyde concentrations in the flue gas increase rapidly with decreasing temperature.<sup>17</sup>

Additional tests<sup>16</sup> on the 250-ton-per-day unit show that formaldehyde emissions increase as underfire air decreases. This trend also was influenced by furnace temperatures.

#### Summary

A source-sampling program has been conducted to determine the pollutant emissions to the atmosphere from many

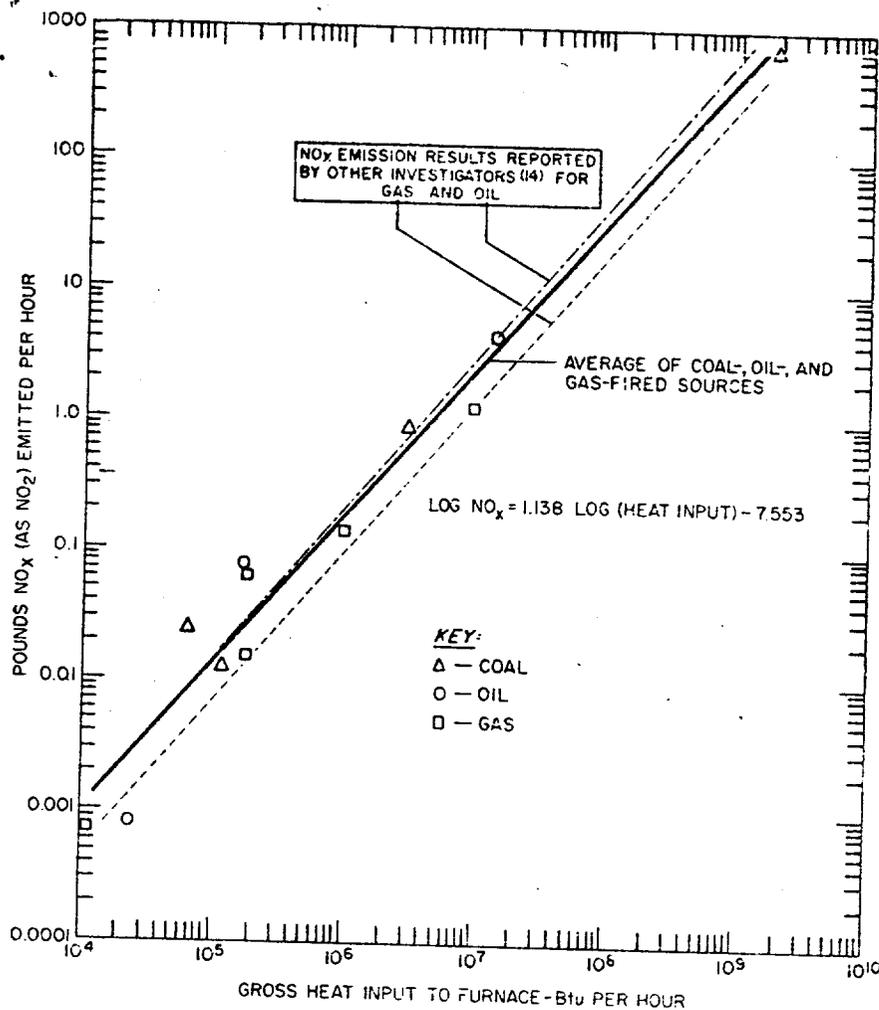


Fig. 6. Relationship of oxides of nitrogen emissions to furnace heat input.

common types of combustion processes. Emission levels of polynuclear hydrocarbons, particulate matter, carbon monoxide, total gaseous hydrocarbons, oxides of nitrogen, oxides of sulfur, and formaldehyde have been measured for (1) heat-generation sources that burn coal, fuel oil, and natural gas and (2) incinerators that burn municipal-type refuse. Also, the polynuclear hydrocarbon concentrations in particulate matter emitted from open fires burning household refuse, automobile tires, grass and hedge clippings, and automobile bodies have been determined. Emission levels of benzo(a)pyrene have been given particular consideration because of the demonstrated carcinogenic activity of this compound.

In the burning of conventional fuels for heat-generation processes or in the disposal of solid waste materials by incineration, the quality or degree of combustion attained is a major factor affecting the release of organic-type pollutants to the atmosphere. Relatively complete combustion of fuel, as measured by the flue-gas composition, is commonly achieved with gas- and oil-fired heat-generating units of any size. A lesser degree of combustion is characteristic of small- and intermediate-

sized coal-fired units and solid-waste-burning incinerators.

The polynuclear hydrocarbon emission rates from coal burning are higher than from oil or gas burning. BaP emission rates for the coal-burning units ranged from 19 to 400,000 micrograms per million Btu gross heat input and decreased as unit size increased. Among the coal-burning units the highest emission rates were produced by a hand-stoked residential coal-fired furnace. A pulverized coal-fired electric power generation plant, the largest source tested, had the lowest polynuclear hydrocarbon emission rates.

BaP emission rates for incinerators ranged from 0.075 to 260 micrograms per pound of refuse charged. The highest emission rates occurred in the small commercial-sized units. Open burning of waste material produced particulate matter containing from 11 to 1100 micrograms of BaP per gram of particulate.

Coal burning and refuse burning are more productive of solid particulates than either fuel oil or gas burning. The extent to which these particulates are formed and released to the atmosphere is dependent on the quality of combustion achieved, the ash con-

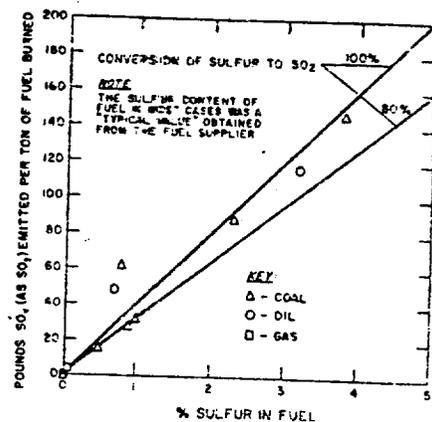


Fig. 7. Sulfur oxides emissions compared with sulfur content of fuel.

tent of the fuel, the type of firing, and whether particulate control equipment is used.

Oxides of nitrogen are produced by the burning of all fuels and waste materials regardless of the quality of the combustion process; these compounds are formed in greater amounts in the larger heat-generating and incineration units. Emission rates for the heat generation units calculated on a pound per hour basis increase as the 1.14th power of the gross heat input to the furnace. Gas-fired units produced lower emissions than did equivalent-sized coal- or oil-fired units.

Oxides of sulfur emissions are a function of the sulfur content in the fuel burned. More than 80% of the sulfur in the fuel was converted to oxides of sulfur.

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