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Publication Date
1996-08-01

LBL-38123
UC-1600
Preprint

ERNEST ORLANDO LAWRENCE BERKELEY NATIONAL LABORATORY

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August 1996
Submitted to
Journal of Air and Waste Management Association

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LBNL-38123
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POLLUTANT EMISSION FACTORS FROM RESIDENTIAL NATURAL GAS APPLIANCES: A LITERATURE REVIEW

by

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August 1996

This work was funded by the California Institute for Energy Efficiency under Agreement No. AQ-92-01B through the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

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Abstract

There is a need to reduce air pollutant emissions in some U.S. urban regions to meet federal and state air quality guidelines. Opportunities exist for reducing pollutant emissions from natural gas appliances in the residential sector. A cost-benefit analysis on various pollutant-reducing strategies is needed to evaluate these opportunities. The effectiveness of these pollutant-reducing strategies (e.g., low-emission burners, energy conservation) can then be ranked among themselves and compared with other pollutant-reducing strategies available for the region. A key step towards conducting a cost-benefit analysis is to collect information on pollutant emissions from existing residential natural gas appliances.

An extensive literature search was conducted to collect data on residential natural-gas-appliance pollutant emission factors. The literature primarily describes laboratory tests and may not reflect actual emission factor distributions in the field. Pollutant emission factors for appliances operated at over 700 test conditions are summarized for nitrogen oxides, carbon monoxide, fine particulate matter, formaldehyde, and methane. The appliances for which pollutant emissions are summarized include forced-air furnaces; stand-alone space heaters (vented and unvented); water heaters; cooking range burners, ovens, and broilers; and pilot lights. The arithmetic means of the nitrogen oxides and fine particulate matter emission factor distributions agree well with the Environmental Protection Agency published emission factor values for domestic gas appliances (in report AP-42). However, the carbon monoxide and methane distribution means are much higher than the relevant AP-42 values. Formaldehyde emission factors are not addressed in AP-42, but the emission factor mean for formaldehyde is comparable to the AP-42 emission factor value for total hydrocarbon emissions.

Introduction

Outdoor air pollution levels fail to meet national ambient air quality standards in many urban areas of the United States. For example, in California's South Coast Air Basin, federal standards for ozone, carbon monoxide (CO), fine particulate matter (PM10, particles less than 10 μm), and nitrogen dioxide (NO_2) standards are not met.[1] The South Coast Air Quality Management District (SCAQMD) is the relevant outdoor air quality agency for the California's South Coast Air Basin. SCAQMD must reduce emissions of nitrogen oxides (NO_x) and volatile organic compounds (VOC) by approximately 80% in the next twenty years to comply with air quality standards according to the Air Quality Management Plan approved by its board of directors in 1989.[1] Such pressures to reduce air pollutant emissions will increase in other urban areas as populations increase and standards become stricter.

It is necessary to know current pollutant emission factors from various sources across the industrial, transportation, commercial, and residential sectors to develop plans for emissions reduction. Although the bulk of pollutant emissions may be from industry and transportation in many areas, commercial and residential contributions to ambient air quality problems can be important.

Opportunities exist for reducing pollutant emissions from the residential sector. The residential sector accounts for 47% of natural gas sales by the U.S. utility industry.[2] Natural gas is considered to be a relatively clean fuel; however, some emissions do occur from its combustion. Two key nitrogen-containing air pollutants produced by natural gas appliances are nitric oxide (NO) and nitrogen dioxide (NO_2); although, recent studies indicate that other nitrogen species may also be produced.[3] Emissions of CO and hydrocarbons are a function of burner design and tuning conditions, and have the potential to become excessive under non-optimal combustion conditions.

The costs and benefits of reducing pollutant emissions from the residential sector need to be addressed and compared against the costs and benefits of pollutant-reducing measures in other sectors. Data on the pollutant emission factors from existing combustion appliances must be gathered prior to conducting such an analysis. Emission factors of residential natural gas appliances have been extracted from existing literature and summarized. The literature primarily describes laboratory tests and may not reflect actual emission factor distributions in the field. Pollutant emission factors reported here are expressed as the mass of pollutant emitted per unit of fuel energy consumed by the appliance.

Methods

Literature Search

A literature search was conducted to find all published quantitative test data reporting pollutant emissions from residential natural gas appliances. The search was conducted by three methods: a computer on-line search, a journal index search, and a search through institution research reports and conference proceedings.

All major technical journals related to natural gas combustion and air pollution were reviewed for appropriate research articles. Keywords used in the search included at least five categories: (1) *general*, e.g., residential, small industrial, and commercial gas appliances; (2) *fuel*, e.g., natural gas and methane; (3) *pollutants*, e.g., carbon monoxide, nitric oxide, nitrogen dioxide, nitrogen oxides, formaldehyde, particulate matter, and associated compounds; (4) *appliances*, e.g., all common household gas appliances such as space heaters, water heaters, and ranges; and (5) *institutions*, e.g., research, utility, and government.

Conference proceedings and technical reports on combustion-related air pollution were also reviewed. Publishers of these reports include the American Gas Association, the Gas Research

Institute, the U. S. Environmental Protection Agency, and the Lawrence Berkeley National Laboratory. In addition, letters were sent to several institutions and researchers to solicit their most recent reports on pollutant emissions from natural gas appliances.

Emission Data Evaluation and Selection

A large number of technical papers and reports were identified through the literature search process. Most of the literature reported laboratory test data on pollutant emissions from domestic natural gas appliances. Only a few researchers have conducted pollutant-emission measurements under actual field conditions. Typically, laboratory tests were designed to simulate a range of potential burner conditions that might be found in the field, e.g., blue flame or yellow flame combustion conditions. Some authors reported test results of domestic appliances in "as found" conditions. Field and laboratory data were used to develop appliance emission factor distributions reported here.

It was not uncommon in the literature to find that a given appliance was tested many times under different combustion and environmental conditions. Examples of the different test conditions cited include: "as found" or "new"; "well-tuned" (blue flame) or "mal-tuned" (yellow flame) burners; different fuel input rates; different air shutter settings; and individual- or multiple-burner tests. Individual test data for each test condition were used in our analysis rather than using average emission data for a given appliance that had been tested under different conditions. Most laboratory test conditions were designed to mimic a potential field condition. Multiple tests of a given appliance operated under the same test condition were averaged before being included in our analysis.

It is important to note that a summary of these data cannot be truly representative of emission factors in the real population of appliances. The data were selected by their availability in the literature, not randomly from the total population of residential appliances. The data represent the appliance selections and operating conditions of various researchers and organizations, and are therefore subject to bias.

The ability of the summarized data to reflect the central tendency of real-world emissions distributions is dependent on the inherent sensitivity of the combustion chemistry to various design and operating parameters. Since NO_x emission factors of conventional burner designs are relatively stable with respect to design and operating parameters, it can be expected that the central tendencies for the summarized distributions of NO_x emission factors would be close to those in the real world. This is less likely to be the case for other products of combustion such as CO or formaldehyde, which vary more because the combustion chemistry leading to the formation of these products is much more sensitive to operating and flame conditions.

Steps were taken to avoid duplication of data collected from different sources. Summary papers and reports where emission data were averaged over a number of appliances were not used. Instead, original emission factor data of individual appliances were used.

The following appliances are included in our analysis: furnaces; stand-alone space heaters; water heaters; cooking ranges; and ovens and broilers. These appliances consume most of the natural gas in residences. High fuel consumption forced-air furnaces dominated the furnace category while lower-output wall heaters and unvented space heaters dominated the space heater category. Pilot light emissions were summarized as a separate category. Gas clothes dryers, pool/spa heaters and other appliances were not included because of the lack of data.

Pollutants included in the analysis are NO_x, CO, CH_4, fine particles (surrogate for PM10), and formaldehyde (HCHO). NO_2 (a component of NO_x), CO, and PM10 are three pollutants that can exceed federal standards in polluted urban air-sheds. NO_x, consisting of NO and NO_2 in this report, are precursors to photochemical smog. CH_4, the main ingredient of natural gas can be emitted in cases of incomplete combustion. Formaldehyde, also a product of incomplete combustion is an irritant, as well as a potential carcinogen and a contributor to photochemical smog formation.

Pollutant emission factors found in the literature are expressed in a variety of units because of a lack of standardization. These units include micrograms per kilo Joule (μg/kJ), micrograms per kilo calorie (μg/kcal), grams per million-calorie (g/10^6 cal), pounds per million-BTU (lb/10^6 BTU), pounds per million-cubic-feet of natural gas (lb/10^6 ft^3), part-per-million air-free flue gas concentration (ppm), and percentages of flue gas concentration. All of these emission factor data were converted into nanograms per Joule (ng/J) for presentation here.

Conversion factors for nitrogen oxides (NO_x) emission factors were not always straightforward. Some authors reported their NO_x emission factor data as NO_2, and some as N. We chose to report our NO_x data as N by using simple molecular weight conversions where appropriate. If the authors reported individual NO and NO_2 measurements, we calculated the total mass (grams) of atomic nitrogen (N) emitted by NO and NO_2 individually and summed the components.

The majority of the appliance emissions measurements of particulate matter (PM) in the literature did not use particle size selection methods with a 10-μm (PM10) cut point. Emission data for particulate matter are typically measured as sub-micron, respirable, or fine particles. These particles have an aerodynamic diameters of 2.5 μm or less. In contrast, the PM10 standard is for particle diameters of 10 μm or less.

Occasionally, it was necessary to infer values for data which were presented as below the limit of detection (LOD). Using inferred values was chosen as the alternative to either ignoring the below-LOD data and creating additional bias, or assuming an arbitrary value of zero for these data points which would not have allowed for geometric summary statistics. Values for these below-LOD data points were inferred using the "robust method of estimating summary statistics."[4] An *a priori* assumption of this method is that the data are log-normally distributed. A brief description of this method follows. Initial values below the lowest reported value are selected at random from a uniform distribution of values between zero and the LOD. The data, both the above LOD and the initially selected below LOD values, are then ranked and normal scores are calculated for each data point. A least-squares linear regression of the logarithm of above-LOD data against their normal scores is conducted. The initial below-LOD data points are then shifted to fit on the calculated regression line. This procedure forces the inferred below-LOD data to fit the log normal distribution defined by the above-LOD data and the number of points below-LOD. The new data, including the fitted below-LOD data, are summarized with arithmetic and geometric means and standard deviations.

Data Summary Methods

The data extracted form the literature were entered into a spreadsheet. The emission data from each individual paper/report was first entered into a file with the original units used by the authors. One data point represented one burner of one appliance operated at one specified condition. These data were checked against the original data to insure the validity of the data entry. All data were then converted to units of ng/J and combined into the emission factor summary file containing data on over seven hundred appliance-combustion conditions. This file was sorted into six appliance/burner types: furnace; space heater; water heater; range burner; oven and broiler; and pilot lights. Again, the furnace category included large whole-house forced-air furnaces while the space heater category included smaller wall furnaces and unvented space heaters.

The database was checked by two people for errors in data entry and conversion calculations. The only reports rejected for this analysis were those that only summarized data of other reports and those that contained zero or below-LOD emission factors for nitrogen oxides. No data from the latter reports were included in our analysis because they appeared to be flawed in either their measurement technique or their methodology since zero NO_x emissions are not possible.

Results

Tables I to V summarize the pollutant emission factors distributions for each of the five pollutants. They include the numbers of the six appliances/burners types collected. Arithmetic and geometric means and standard deviations for the pollutant emission factor distributions are presented. Figures 1 to 5 show these emission factor frequency distributions for NO_X (as N), CO, fine particles, HCHO, and CH_4. For comparison, the AP-42 emission factor values for domestic and commercial boilers are also shown in the relevant figure.[5]

Nitrogen Oxides (NO_x) Emission Factors

Figure 1 shows the distributions of nitrogen oxides (as N) emissions for 555 data points summarized from the literature.[6-26] The simple average of all five appliance types (excluding the pilot light average emission factor) is 12.7 ng/J, which agrees closely to the AP-42 value of 12.5 ng/J after conversion from NO_X (as NO_2) to NO_x (as N). The AP-42 value is currently used by the SCAQMD as the NO_X water heater emission standard. As expected, the distribution of NO_X emission factors is narrow, with geometric standard deviations (GSDs) ranging from 1.3 to 1.4 for the different appliances.

Data were also available on the individual components of NO_x for 522 burner conditions. On average, 32% (± 18%) of the nitrogen in NO_x was in the form of NO_2 and the remainder was in the form of NO. The corresponding geometric mean (GM) of the ratio of N (in NO_2) divided by N (in NO_x) was 26% (GSD = 2.22). There was a noticeable difference in this ratio for burners tested under blue-flame and yellow-flame conditions. For 231 blue-flame conditions, the N (in NO_2) portion of N (in NO_x) had an arithmetic mean (AM) of 25% (± 13%) and a GM of 20% (GSD = 2.36). For 149 yellow-flame conditions, the AM was 37% (± 20%) and the GM was

30% (GSD = 2.22). These results are consistent with previous work that showed the NO_2/NO_x ratio rises under excess combustion-air (yellow-flame) conditions although the total NO_x emissions fall.[24] Data collected here showed that NO_x (as N) emission factors are about 0.6 ng/J lower for yellow-flame burners compared with blue-flame burners.

Carbon Monoxide (CO) Emission Factors

Figure 2 shows the distributions of carbon monoxide emission factors for the different gas appliances summarized from the literature.[7-16,18-27] The data are plotted on a semi-log scale and resemble a bi-modal distribution. A total of 661 burners or burner-conditions are included in the analysis. The simple average emission factor for the 5 appliance types (excluding the pilot light emissions) is 72.6 ng/J; almost 9 times higher than the 8.2 ng/J value published in AP-42. The emission factors range over 3 orders of magnitude. The high end of the bi-modal distribution for all burners (see Figure 2) appears to reflect the over representation of high-CO-emitting range burners and a large number of burners tested under yellow-flame (mal-tuned) conditions.

Fine Particulate Matter Emission Factors

Figure 3 shows the distributions of particulate matter emission factor measurements from furnaces, space heaters, range burners, and ovens and broilers found in the literature.[13,22-24,28] These tests measured sub-micron or fine particle emissions rather than standardized PM10 emissions. However, particles emitted from natural gas combustion are predominately less than 1 μm in aerodynamic diameter,[22] thus these emission factors can be considered as a surrogate for PM10 emission factors. The total of 41 appliance burners tested for particle emissions are reported. The simple four-appliance arithmetic mean of 0.5 ng/J agree with the AP-42 value for PM10, which is between 0.4 and 2.1 ng/J. Only one test each were found in the literature for range burner and oven particle emissions; both were below 0.5 ng/J. No particulate emissions data were found for natural gas water heaters or pilot lights.

The particulate matter emission factor data for space heaters included 17 values which were reported as below the LOD. The summary statistics for these 17 data points include inferred values for these low emission factors calculated using the method discussed above.

Formaldehyde (HCHO) Emissions

Figure 4 shows the distributions of formaldehyde emission factors for 70 burner-conditions.[10,12,13,16,20,23,24] The simple mean emission factor for the four appliances is 0.9 ng/J. Although there is no AP-42 value for formaldehyde, the AP-42 value for all hydrocarbon emissions is only slightly higher (1.1 ng./J) than our appliance mean.

Methane (CH_4) Emissions

Figure 5 shows the distributions of methane emissions from gas 90 burners or burner conditions.[14,19,20,26] The simple appliance arithmetic mean emission factor for CH_4 is 6.7 ng/J, a factor of 6 larger than the AP-42 value of 1.1 ng/J for total hydrocarbon emissions. The highest emission factors were for pilot lights averaging 77 ng/J.

The CH_4 emission factor data for water heaters and space heaters included 8 and 2 values, respectively, which were below LOD. The summary statistics for these data include inferred values for these low emission factors calculated using the method discussed above.

Conclusions

Pollutant emission factors from residential natural gas appliances reported in the literature have been summarized. These summaries can be used as input to models assessing total pollutant emissions from natural gas use in the residential sector.

The mean values for the distributions of emission factors for nitrogen oxides and fine particulate matter are in good agreement with the AP-42 values published by the Environmental Protection

Agency. However, the mean values for carbon monoxide emissions are much higher than the AP-42 values. The methane emission factors for pilot lights and space heaters are also much higher that the AP-42 value for total hydrocarbons.

These results indicate that attempts to estimate residential stationary source emissions based on the AP-42 values alone may underestimate the contribution of natural gas appliances to ambient levels of CO and total hydrocarbons. Care must be taken in interpretation of these data since they do not represent a statistically-valid sample of the population of appliances in use. Any biases in the appliances selected by the various studies have likely skewed the summarized results.

Acknowledgments

This work was funded by the California Institute for Energy Efficiency under Agreement No. AQ-92-01B and through the U.S. Department of Energy under Contract No. DE-AC03-76SF00098. The authors thank Jon Koomey and David Faulkner of the Lawrence Berkeley National Laboratory for their reviews of the manuscript.

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Table I. Summary of NO_x (as N) Emission Factors for Residential Natural Gas Appliances.[a]

Appliance Type	Number of Appliances/ Burners	Arithmetic Mean (ng/J)	Arithmetic S.D.(ng/J)	Geometric Mean (ng/J)	Geometric S.D.
Furnace	27	13.5	3.9	12.9	1.37
Space Heater	81	13.3	4.8	12.5	1.41
Water Heater	54	16.1	3.5	15.7	1.27
Range Burner	282	10.1	2.7	9.7	1.29
Oven & Broiler	77	10.5	4.2	9.8	1.41
Pilot Light	34	8.0	2.2	7.6	1.39
Simple Appliance Average (w/o pilot)		12.7			
AP-42		12.5			

[a] Summarized from references 6-26.

Table II. Summary of CO Emission Factors for Residential Natural Gas Appliances.[a]

Appliance Type	Number of Appliances/ Burners	Arithmetic Mean (ng/J)	Arithmetic S.D.(ng/J)	Geometric Mean (ng/J)	Geometric S.D.
Furnace	16	31.0	38.7	11.8	4.96
Space Heater	81	50.1	95.2	15.1	4.92
Water Heater	55	12.2	19.6	5.1	3.65
Range Burner	369	165	216	83.0	3.50
Oven & Broiler	102	104	148	45.9	3.80
Pilot	38	89.8	102.3	58.5	2.42
Simple Appliance Average (w/o pilot)		72.6			
AP-42		8.2			

[a] Summarized from references 7-16, 18-27.

Table III. Summary of Fine Particle Emission Factors for Residential Natural Gas Appliances.[a]

Appliance Type	Number of Appliances/ Burners	Arithmetic Mean (ng/J)	Arithmetic S.D.(ng/J)	Geometric Mean (ng/J)	Geometric S.D.
Furnace	4	1.12	1.92	0.29	6.73
Space Heater[b]	35	0.38	1.18	0.01	25.5
Range Burner	1	0.41		0.41	
Oven & Broiler	1	0.05		0.05	
Simple Appliance Average (w/o pilot)		0.49			
AP-42		0.4-2.0			

[a] Summarized from references 13, 22, 23, 24, and 28
[b] Seventeen emission factors were inferred from values reported as below limit of detection.

Table IV. Summary of HCHO Emission Factors for Residential Natural Gas Appliances.[a]

Appliance Type	Number of Appliances/ Burners	Arithmetic Mean (ng/J)	Arithmetic S.D.(ng/J)	Geometric Mean (ng/J)	Geometric S.D.
Furnace	7	0.34	0.60	0.14	3.59
Space Heater	49	1.17	2.96	0.35	5.23
Range Burner	11	0.68	0.46	0.53	2.18
Oven & Broiler	3	1.48	1.27	0.93	4.05
Simple Appliance Average (w/o pilot)		0.92			
AP-42		n.a.			

[a] Summarized from references 10, 12, 13, 16, 20, 23, and 24.

Table V. Summary of CH_4 Emission Factors for Residential Natural Gas Appliances.[a]

Appliance Type	Number of Appliances/ Burners	Arithmetic Mean (ng/J)	Arithmetic S.D.(ng/J)	Geometric Mean (ng/J)	Geometric S.D.
Furnace	2	0.98	1.15	0.55	5.35
Space Heater[b]	35	18.4	48.7	2.14	8.12
Water Heater[c]	32	0.70	0.64	0.47	2.62
Pilot	21	77.0	157.0	13.1	6.42
Simple Appliance Average (w/o pilot)		6.7			
AP-42[d]		1.1			

[a] Summarized from references 14, 19, 20, and 26.
[b] Two emission factors were inferred from values reported as below limit of detection.
[c] Eight emission factors were inferred from values reported as below limit of detection.
[d] For total hydrocarbon emissions.

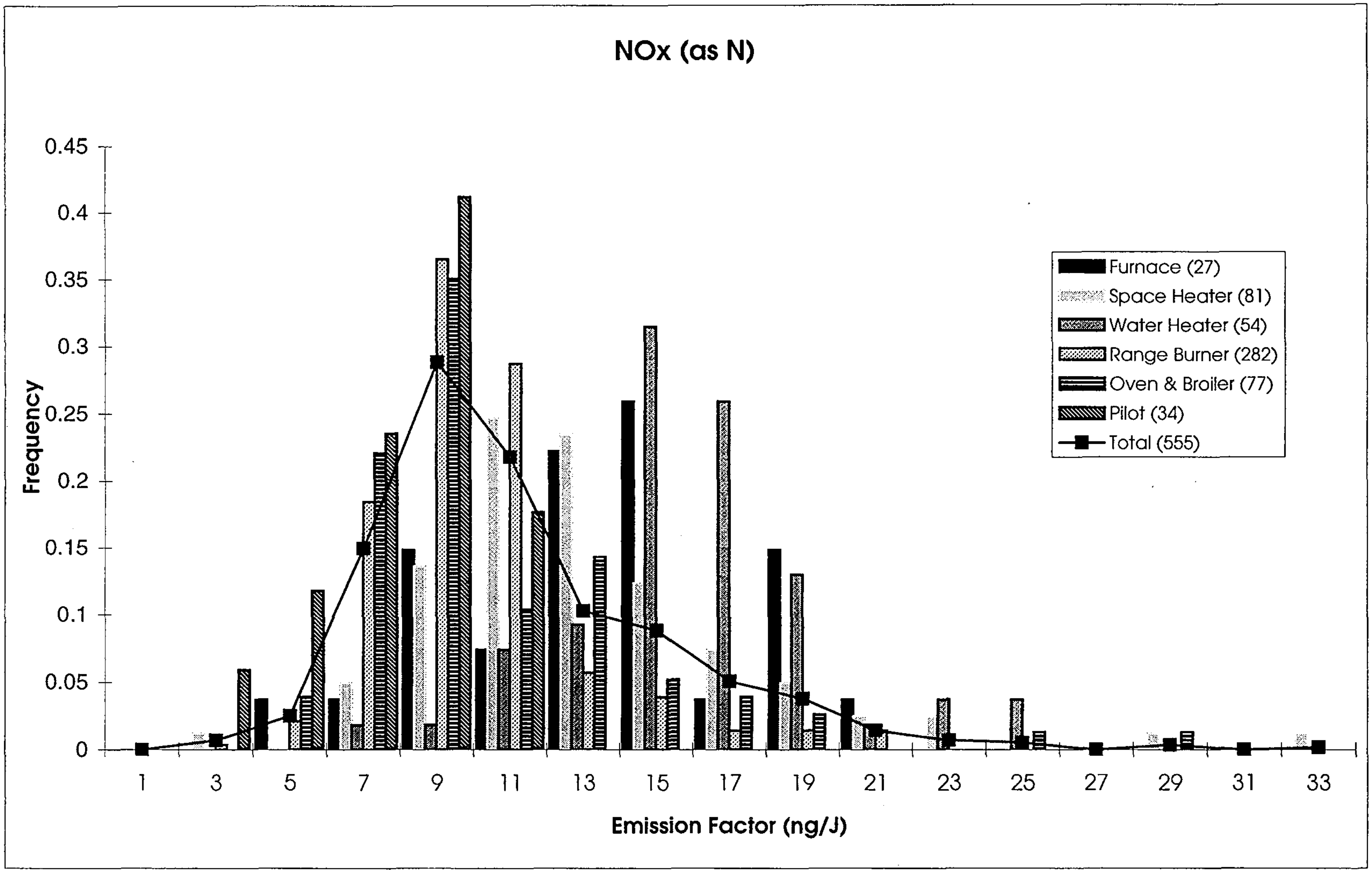

Figure 1. Nitrogen oxides as nitrogen [NOx (as N) emission factor (ng/J) distributions from residential natural gas appliances.

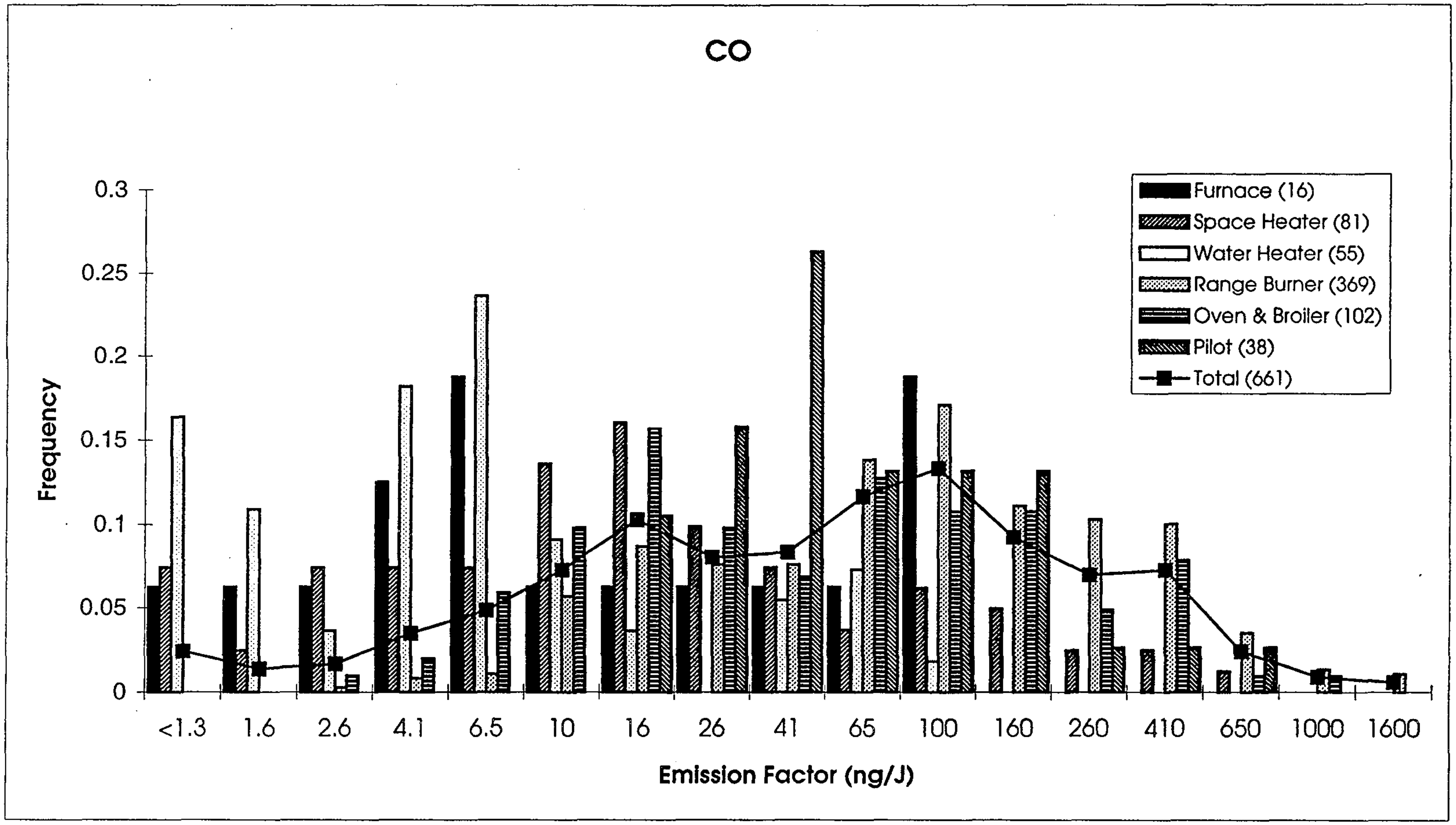

Figure 2. Carbon monoxide (CO) emission factor (ng/J) distributions from natural gas appliance burners.

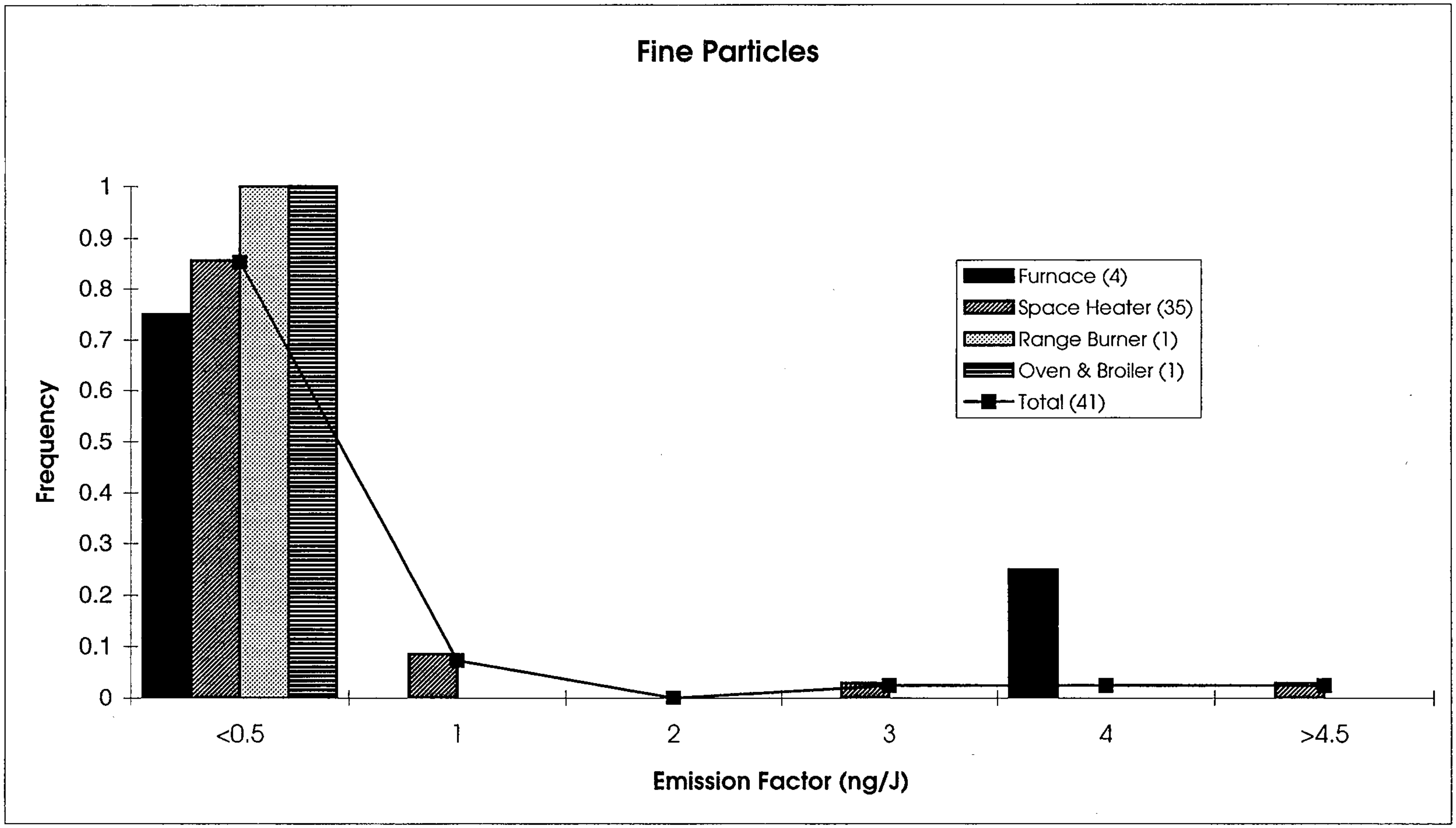

Figure 3. Fine particle emission factor (ng/J) distributions from residential natural gas appliances.

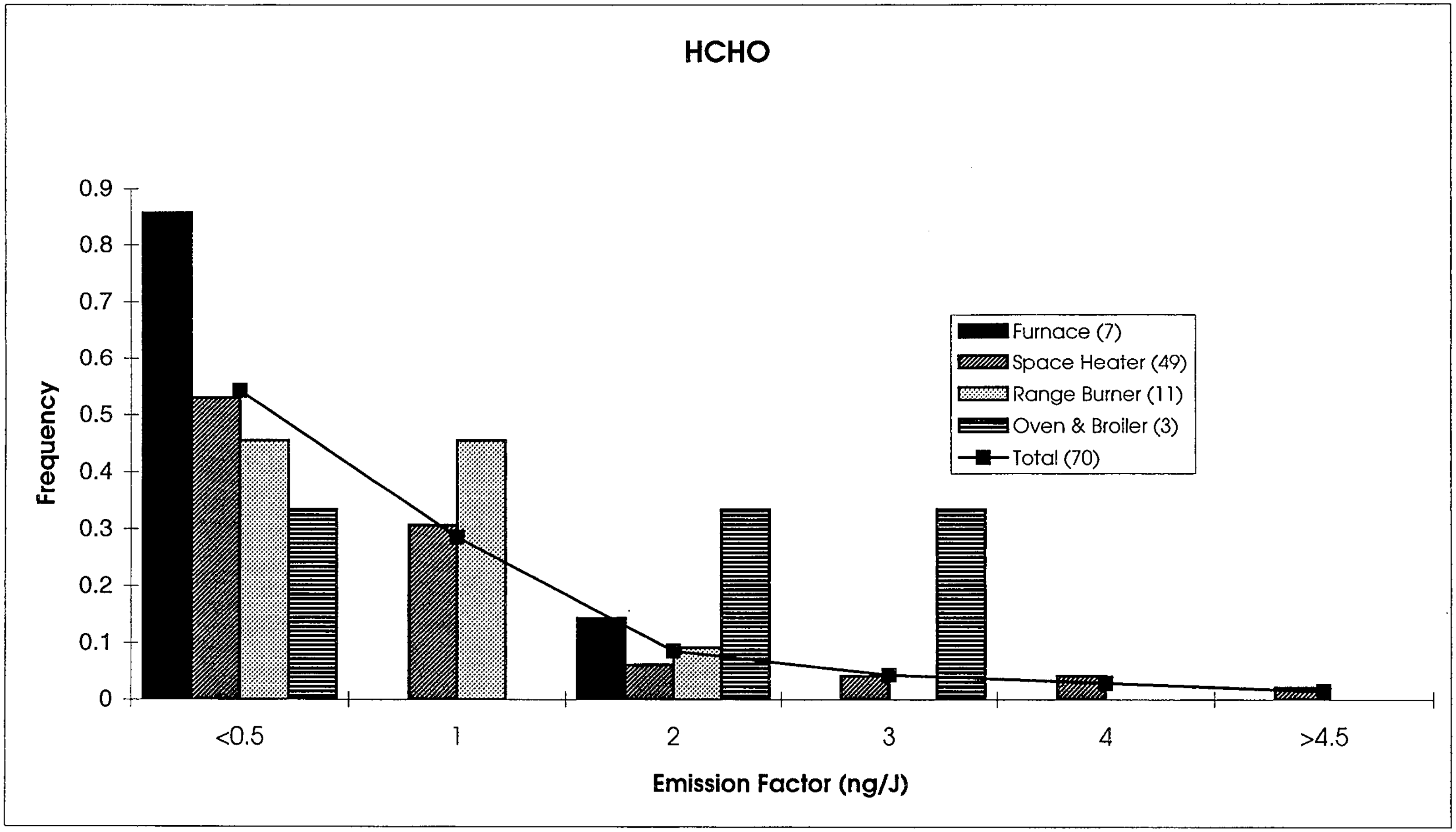

Figure 4. Formaldehyde (HCHO) emission factor (ng/J) distributions from residential natural gas appliances.

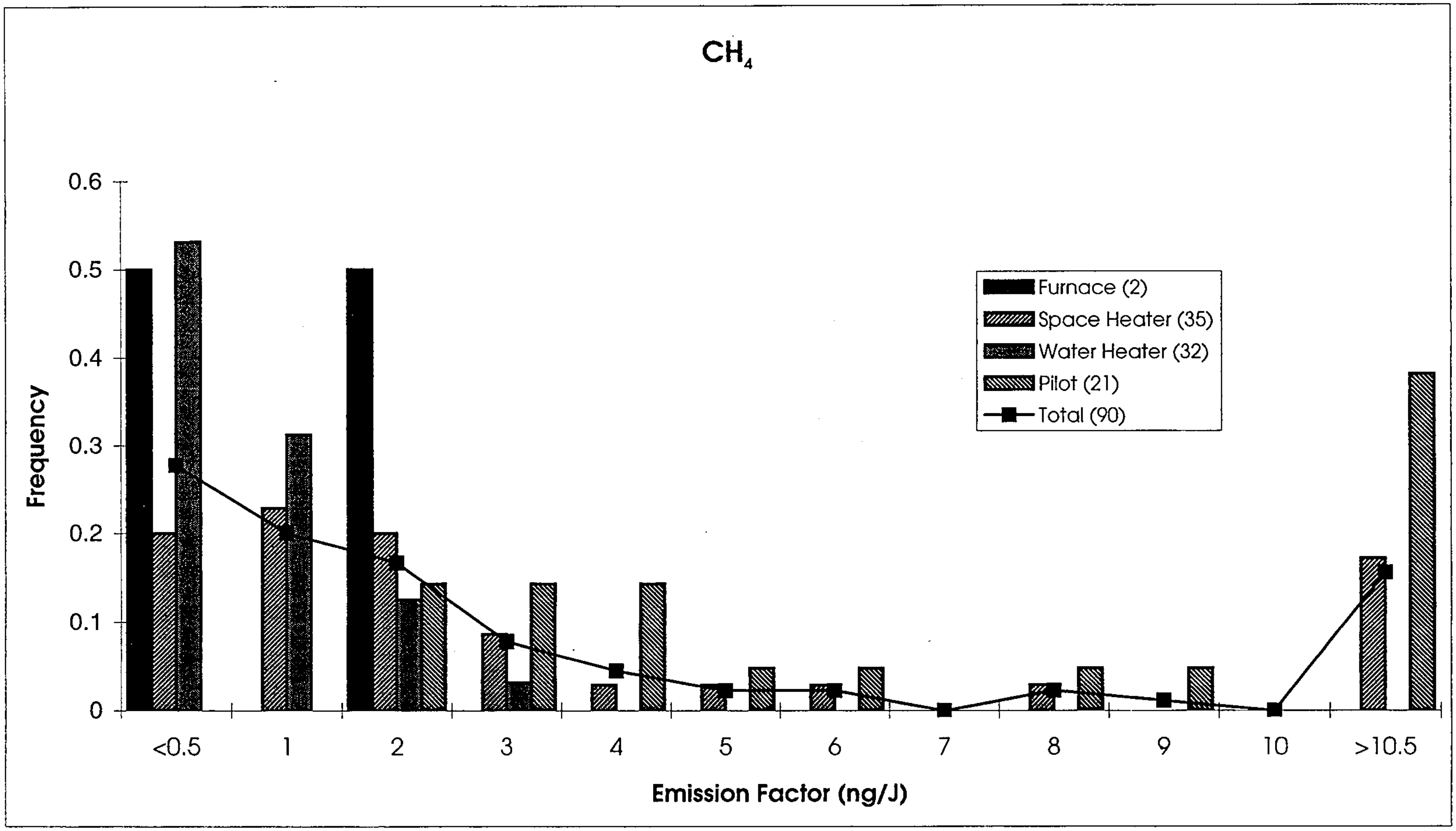

Figure 5. Methane (CH4) emission factor (ng/J) distribution from residential natural gas appliances.