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# NITROGEN OXIDE EMISSIONS FROM A PILOT PLANT

## SPREADER STOKER BARK FIRED BOILER

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RICHARD ALLEN KESTER

A dissertation submitted in partial fulfillment  
of the requirements for the degree of

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UNIVERSITY OF WISCONSIN

1980

Approved by

Department

Signature

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*[Handwritten Signature]*  
*[Handwritten Signature]*

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by

RICHARD ALLEN KESTER

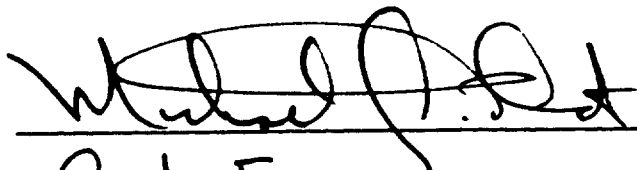
A dissertation submitted in partial fulfillment  
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UNIVERSITY OF WASHINGTON

1980

Approved by



Department

Civil Engineering

Date

December 17, 1979

DOCTORAL DISSERTATION

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Date: November 30, 1979

We have carefully read the dissertation entitled Nitrogen Oxide Emissions from a Pilot Plant Spreader Stoker Bark Fired Boiler

Richard Allen Kester

submitted by

in partial fulfillment of

the requirements of the degree of Doctor of Philosophy

and recommend its acceptance. In support of this recommendation we present the following joint statement of evaluation to be filed with the dissertation.

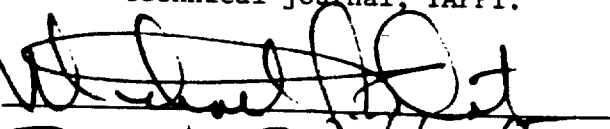
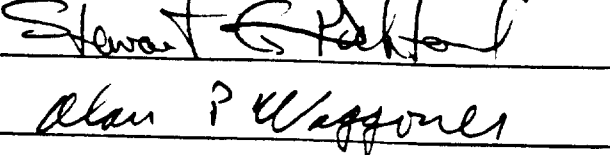
The dissertation involves an experimental study of the emission of nitrogen oxide air pollutants from the combustion of Douglas fir bark in a pilot plant spreader stoker boiler and a correlation of the NO emissions with fuel and boiler parameters.

The dissertation includes:

1. A background section describing the use of wood waste as a fuel and presenting a review of the properties of the nitrogen oxides and a review of the formation of nitrogen oxides in a flame and in wood combustion.
2. A description of the experimental test facility located at Oregon State University and of the methods for the measurement of nitric oxide (NO) and of nitrogen dioxide (NO<sub>2</sub>) in the emission gases, of the fuel feed rate, of the fuel size, of the fuel moisture content, of the excess air for combustion, of the combustion exhaust gas temperature, and of the total underfire air flowrate.
3. The measured NO concentration data was correlated to the independent test parameters including fuel feed rate, fuel moisture content, combustion excess air flowrate, and total underfire air flowrate. These correlations of the nitric oxide concentration in the exhaust gases with the various parameters is new information relating NO emissions to boiler parameters.
4. A new emission factor for use in performing estimation calculations of the NO emissions from hog-fuel boilers was developed. This new NO emission factor <sup>x</sup> is about 11% of the magnitude of the old emission factor (new <sup>x</sup> emission factor is 1.1 pounds of NO as NO<sub>2</sub> per ton of wet bark burned compared to the old factor of 10 pounds of NO<sub>x</sub> as NO<sub>2</sub> per ton of wet bark burned).
5. A new predictive model which enables the NO concentrations to be calculated as a function of the boiler combustion parameters was developed. This predictive model is based upon a linear regression analysis of the data measured during this study.

This PhD dissertation research effort is of considerable significance to the air resources field with respect to: the atmospheric NO<sub>x</sub> concentration to emission source relationship, the control strategies used by air pollution control agencies to reduce ambient air NO<sub>x</sub> concentrations, wood products industries utilization of wood waste as an energy source, and the general use of wood waste as an energy source. This research represents a significant contribution to the Air Resources Engineering knowledge and has already resulted in one publication in a refereed technical journal, TAPPI.

DISSERTATION READING COMMITTEE:

  
Stewart G. Parker  
  
Alan P. Waggoner

UNIVERSITY OF WASHINGTON

ABSTRACT

NITROGEN OXIDE EMISSIONS FROM A PILOT PLANT  
SPREADER STOKER BARK FIRED BOILER

by Richard Allen Kester

Chairman of Supervisory Committee:

Professor Michael J. Pilat

Department of Civil Engineering

The emission of nitrogen oxides, both NO and NO<sub>2</sub> were measured from a pilot plant spreader stoker boiler using Douglas Fir (*Psuedotsuga menziesii*) bark for fuel. The boiler was fired under a wide range of fuel feed rates, excess air rates, fuel sizes, fuel moisture contents, and levels of underfire air. Ninety-seven complete data sets were recorded.

A data base for nitric oxide emissions was developed. Values ranged from 25 to 103 ppm on a dry basis, and corrected to 12 percent carbon dioxide. The average for all tests was 57 ppm and the geometric mean was 56 ppm.

An emission factor in terms of pounds of  $\text{NO}_x$  (calculated as  $\text{NO}_2$ ) emitted per ton of wet bark burned was calculated to be 1.1 pounds of  $\text{NO}_x$  per ton of bark. This is substantially lower than the current EPA emission standard of 10.0 pounds of  $\text{NO}_2$  per ton of wood waste burned.

A predictive model of NO emissions as a function of boiler variables was developed using multiple linear regression analysis techniques. The best fit equation is  $\text{NO (ppm)} = 0.0623 \text{ RATE} + 0.0935 \text{ TEMP} + 0.259 \text{ XAIR} - 6.29 \text{ SIZE} + 60.5 \text{ H}_2\text{O} + 0.678 \text{ UAIR} - 145$ . The equation has a multiple correlation coefficient of 0.86.

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Finally, I want to thank my family, my wife Diane, my son Jason, and my daughter Lisa, for their patience and understanding during these last few years. I realize how difficult it was on them at times when "Daddy has to study tonight."

## I. INTRODUCTION

### A. OBJECTIVES OF RESEARCH

A research project was conducted to investigate the formation of nitrogen oxides ( $\text{NO}_x$ ) in a scale model industrial type spreader stoker hog fuel boiler burning Douglas fir bark, operating over a wide range of combustion variables commonly found in industrial applications. The objectives of the proposed research were to:

1. Develop a data base of the emissions of  $\text{NO}_x$ .
2. Develop an emission factor for use in estimating current and future  $\text{NO}_x$  emissions.
3. Develop a predictive model of  $\text{NO}_x$  generation from wood waste combustion based on boiler operating parameters.

### B. SIGNIFICANCE OF RESEARCH

There are currently no available data which can be used to determine the emissions of nitrogen oxides from hog fuel fired boilers under varying operating conditions. At present very little published data exist on nitrogen oxides from hog fuel fired boilers. It is not known if hog fuel boilers are an important

source of this pollutant, nor how much is being emitted from existing ones.

Wood waste combustion will increase as an energy source as the price of fossil fuel increases, and new, larger, higher temperature and pressure boilers replace older, less efficient units. New air pollution regulations have been developed that require limits on nitrogen oxides emissions from hog fuel boilers. It is desirable to know how much nitrogen oxides are being generated and what factors influence their generation in order to minimize nitrogen oxides if necessary in the design of future combustors.

The current Environmental Protection Agency (EPA) New Source Performance Standard for nitrogen oxides emissions from coal combustion is 0.72 pound per  $10^6$  Btu, and the EPA emission factor for nitrogen dioxide ( $\text{NO}_2$ ) from wood combustion converts to 1.1 pounds per  $10^6$  Btu. Additionally, the EPA emission factor for coal combustion in a spreader stoker is 15 pounds per ton, and wood is rated at 10 pounds per ton. While this indicates that wood will produce somewhat less nitrogen oxides pound for pound than coal, it must be remembered that since wood has a lower Btu content per pound, the total emission of nitrogen oxides from hog

fuel will be greater for a given amount of energy, according to the EPA emission factors.

Using EPA's emission factor, and comparing wood with residual oil, indicates that wood emits almost three times the amount of  $\text{NO}_x$  on a pound of  $\text{NO}_x$  per Btu basis (1.1 pounds per  $10^6$  Btu for wood versus 0.40 pound per  $10^6$  Btu for oil). Because of this, in an area like the Northwest with a significant portion of industrial energy supplied by wood, and with wood steadily replacing fossil fuels, it seems necessary to determine the exact nature and magnitude of nitrogen oxides generation from wood fuels. Table 1 shows  $\text{NO}_x$  emissions from industrial fuels in the Puget Sound area as calculated by the Puget Sound Air Pollution Control Agency.

Table 1. INDUSTRIAL FUEL USE AND EMISSIONS TOTALS KING, PIERCE, SNOHOMISH, AND KITSAP COUNTIES 1975 DATA

<u>Fuel Type</u>	<u>Oil</u>	<u>Natural Gas</u>	<u>Hog Fuel (wet)</u>
Amount	$6.83 \times 10^7$ gal	$2.28 \times 10^8$ therms	$2.85 \times 10^9$ lb
Btu (a)	$1.02 \times 10^{13}$	$2.33 \times 10^{13}$	$1.28 \times 10^{13}$
Percent Btu	22	49.4	27.7
Percent $\text{NO}_x$ Emissions (b)	22	12.8	62.5

- (a) Using  $1.5 \times 10^{13}$  Btu/gal for oil,  $1 \times 10^5$  Btu/therm for gas,  $4.5 \times 10^4$  Btu/lb for wet hog fuel.  
 (b) Using EPA emission factors.  
 (c) Coal supplies the remaining Btu's and  $\text{NO}_x$ .

A significant use of the results of this research will be in having available an emission factor for the amount of  $\text{NO}_x$  emitted per ton of hog fuel burned that is based on actual data. In researching the current EPA emission factor's development, it was learned that the emission factor for hog fuel was assumed to be similar to that for coal, because they were both solid fuels. That level of accuracy may have sufficed during the earlier days of air pollution control, but a more accurate figure is needed now because of the great importance of emission factors used in modeling studies required for new source permits by EPA.

The 1977 Clean Air Act Amendments by Congress provides air quality designations for the entire nation. These are called class I, class II, and class III areas. Areas that have been designated class I include national parks and wilderness areas. It is desired to maintain the existing high levels of pristine air quality in these areas. The rest of the nation is now classified as class II where some increases in air pollution will be allowed. Areas could be designated as class III which now meet the ambient standards, and new growth would be welcomed. Air quality would be allowed to deteriorate up to the level of the secondary ambient standards in these areas.

In these class I, II, and III areas, the EPA has developed increments of air quality degradation that are allowable for a new source being built in the area. For example, the class I increment for particulates is 10 micrograms per cubic meter, class II is 37 micrograms per cubic meter, and class III is 75 micrograms per cubic meter. These are all 24-hour averages. If a new source is constructed in one of these areas, the effect on the air quality must be less than this increment, and certain other criteria must be met. The effect of the proposed source on the allowable increment is determined by the use of ambient air quality models.

The general equation for ambient air contaminant concentration at the centerline of a plume is given as: (Turner, 1967)

$$x = \frac{Q}{(\sigma_y)(\sigma_z)(\pi)(V)} \exp\left[-\frac{1}{2}\left[\frac{H}{\sigma_z}\right]^2\right]$$

Where:  $x$  = ambient air contaminant concentration

$Q$  = emission rate

$\sigma_y, \sigma_z$  = values for dispersion in the  
y and z direction

$V$  = wind speed

$\pi$  = 3.1416

$H$  = plume height

It is obvious that the emission factor used to determine the emission rate (Q) can play a major role in determining if EPA will grant approval for the location of a new source in a specific area.

Another significant result of this research will be the identification of which hog fuel boiler parameters are most closely correlated with NO<sub>x</sub> emissions. This should allow boiler operators to make changes in boiler operation if desired, to reduce emissions of NO<sub>x</sub>.

C. RESEARCH APPROACH

The approach to the proposed research was experimental in nature. The research objectives required that a research facility be identified that had the following characteristics:

1. Of a spreader stoker industrial design and operating exclusively on wood waste.
2. Of a size large enough to simulate industrial boiler operation and small enough not to need enormous quantities of fuel to operate. This ruled out both bench scale operations and most industrial boilers.

3. Flexible enough in operation to be able to tolerate many startups, shutdowns, change in steam rate, changes in fuel, and changes in scheduled operation. This ruled out industrial boilers tied to a process.
4. Located within a reasonable travel distance of the University of Washington.
5. Instrumented or capable of being instrumented to measure combustion parameters.
6. Manned with a crew capable of firing and maintaining the boiler.

The chosen research location was the Fairplay Experimental Wood Combustion Test Facility at Oregon State University in Corvallis, Oregon.

In the summer of 1976, construction began on the wood residue combustion test facility in Corvallis. Construction continued until the end of January 1977, at which time the facility underwent its initial firing. Modifications followed for two months, and initial testing began in April 1977. Those experiments were on wood combustion and the effect of under

and over fire air distribution on particulate emissions by Junge (1977) and an investigation into wood combustion occurring in the burning fuel bed by Tuttle (1978). In the summer of 1978, the Weyerhaeuser Company joined OSU in a joint research effort at the facility to investigate improved combustion technology. It was during these investigations that the author was fortunate enough to use the facility in conjunction with the Weyerhaeuser Company to investigate the emissions of nitrogen oxides from wood combustion.

The experimental boiler test was designed to provide data on  $\text{NO}_x$  emissions over three levels of total air-to-fuel ratios, three levels of under fire air to fuel ratios, a range of fuel moisture contents, fuel of two different sizes, and a wide range of fuel feed rates. Combustion temperature and  $\text{NO}_x$  were measured as dependent variables. Combustion air temperature was held constant at 280 degrees F for each run, and the fuel used is Douglas fir bark for all tests. Table 2 summarizes the range of test parameters.

Table 2. RANGE OF TEST PARAMETERS

Variable	Range
Fuel feed rate	450 to 750 dry pounds per hour
Excess air	30 to 80 percent
Fuel size	0.3 inch (fine), and 0.3 to 1.3 inches (coarse)
Fuel moisture	36.9 to 53.9 percent (wet basis)
Total under fire air	0.9 to 6.5 pounds of air per pound of fuel

An important difference in this study from other bench scale studies on emissions from wood combustion (Prakash, 1972) is that this one was operated in a steady state mode. This eliminated the influence of end conditions on the test data due to cold startup and incomplete total combustion, and should make the results of the experiment more directly applicable to industrial situations.

In order to develop an overall relationship between the measured parameters and NO concentration, it is possible to observe the relationship of individual parameters and from these determine an overall form of the relationship. However, because of the complexity of deriving a general equation to cover all the variables, a multiple linear regression analysis (Stat Pack V4, Western Michigan University) was used, and

a linear equation predicting NO values based on the .  
measured combustion parameters was developed.

## II. BACKGROUND

### A. WOOD WASTE AS A FUEL

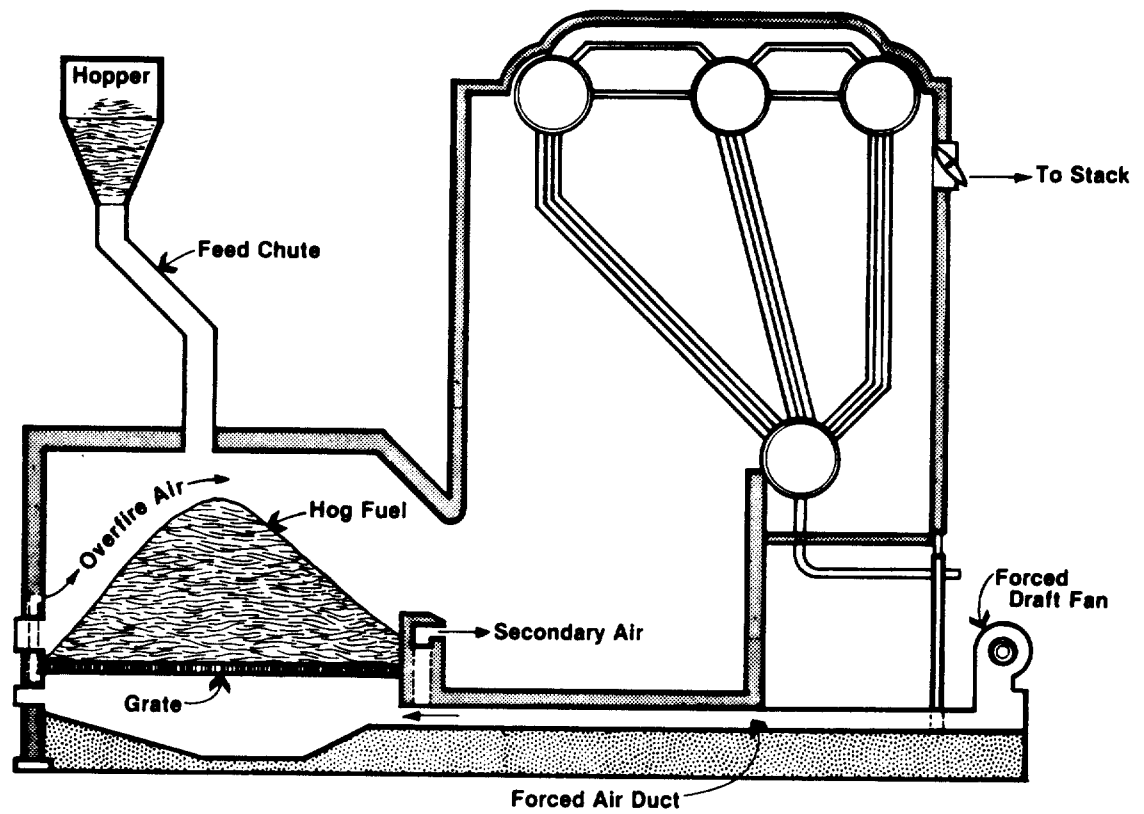
The industrial use of wood as a fuel has been practiced for many decades in this country. In the early part of the century, wood was a major source of energy but was gradually replaced by coal and, later, by petroleum resources. Today, wood fuel is limited primarily to use in the forest products industries. There, the residue from logging practices and from wood processing operations is burned to supply process steam and electricity. Wood residue is an important fuel to this industry. Energy derived from wood residue in industrial boilers in 1977 in Washington State was equivalent to over 200 million gallons of oil according to the Washington Department of Ecology (1977). There are approximately 1,000 industrial wood residue fired boilers in operation in the Pacific Northwest (Junge, 1977).

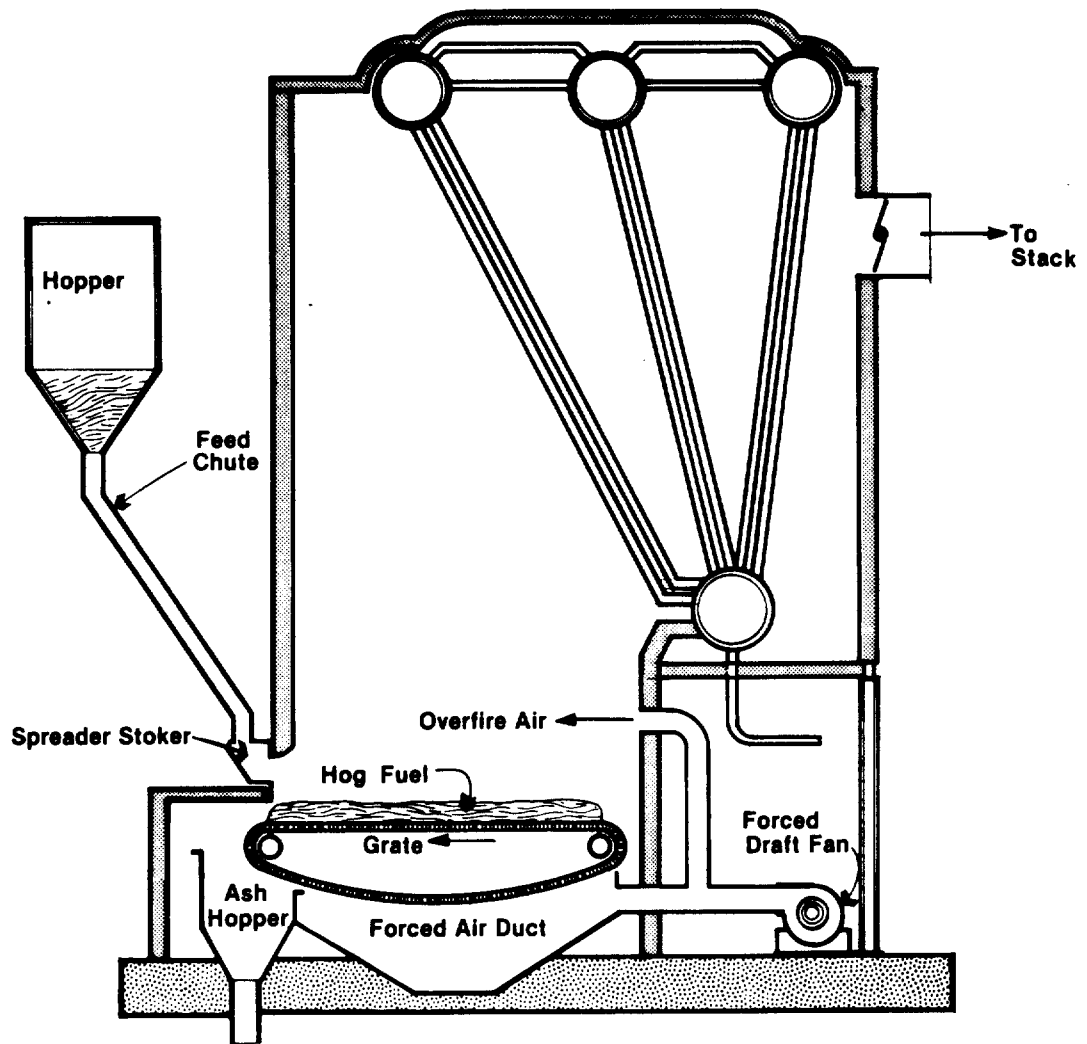
Wood residue which is burned in these industrial boilers is usually reduced in size in a machine called a "hog" before it is sent to the boilers. Because the fuel is sent through a hog, it is generally referred to as "hogged fuel" or "hog fuel." It may

consist of bark, planer shavings, sawdust, sander dust, slabs, trim from plywood, particleboard, and hardboard operations, and miscellaneous other wood residue materials. In the Northwest, the fuel is usually a mixture of bark and unusable white wood, with the majority of the fuel being bark. Much of the hog fuel is burned in the natural "wet" condition and may contain 50 percent or more water by weight.

The boilers used to burn this hog fuel have seen a gradual evolution from the original designs of the late 19th century. In those early units, the fuel was dropped in a large mound on the grates of a refractory-lined chamber called a Dutch Oven as shown in figure 1. Distillation of the water and volatiles along with initial combustion took place in the Dutch Oven. The completion of combustion took place in an adjoining conventional boiler furnace. Steam-generating boilers of this type are called Dutch Oven boilers.

In the late 1940's, the Dutch Oven design began to be replaced by a combustion design called the spreader stoker as shown in figure 2. In this design, the fuel is spread across a conventional coal boiler-type grate in a thin layer using a mechanical or pneumatic spreader, and the Dutch Oven is eliminated. Spreader stoker

**Dutch Oven Boiler****FIGURE 1**



**Spreader Stoker With Traveling Grate**

**FIGURE 2**

boilers are usually equipped with water wall tubes and have much less refractory than the Dutch Oven boilers. They are more responsive to changes in steam demand because of the smaller mass of fuel being burned at any instant and are less expensive to construct and maintain.

A third type of combustion system is the suspension burner in which the hog fuel is reduced to a small size and burned in suspension rather than on top of a grate. Fuel cells are commonly of the suspension burning type. They include both vertical and horizontal cylindrical combustion chambers. Combustion temperatures in these units are generally the highest of the three types of combustion systems.

At present, most hog fuel boilers are Dutch Oven or spreader stoker types. Both have relatively low combustion temperatures. With the increasing value of wood for energy, larger and more efficient furnaces are replacing existing units. There is also an increasing interest in the use of suspension burning combustors and precombustion fuel dryers. These new boilers will supply more power than the old ones and may operate at substantially higher combustion temperatures. Since nitrogen oxide formation is reported

to be directly related to combustion temperature (Zeldovich, 1946), the future emissions of nitrogen oxides from wood combustion may be far in excess of those encountered today.

#### B. PREVIOUS RESEARCH

Observing and reporting on nitrogen oxide formation from fossil fuel combustion has become a popular activity in recent years. It has been studied in depth by many authors, and the proposed mechanisms and rates of reactions of nitrogen oxide formation have been reported for these fuels over a wide range of variables (Bagwell, 1971; Engleman, 1973; Fenimore, 1971, 1972, 1976; Flagan, 1974; Kramlich, 1978; Malte, 1974, 1977, 1978; Wendt, 1977; Westernberg, 1971; Zeldovich, 1946).

Wood combustion and air contaminant emissions have also been studied (Boubel, 1958, 1965; Corder, 1965, 1970, 1973; Droege, 1965; Junge, 1977; Kester, 1972; Kreisinger, 1939; Prakash, 1972), and similar types of findings for combustion reaction rates and combustion contaminants (other than nitrogen oxides) have been reported. However, a review of the literature has failed to produce articles on nitrogen oxides

concentrations in hog fuel boiler flue gas other than brief remarks by Galeano (1973), Schmall (1978), and Hunter (1979).

Galeano reported a value of 123 ppm in a paper on nitrogen oxide emissions from a kraft pulp mill with the indication that the amount of natural gas being burned with the hog fuel was very small. Schmall reported a value of 55 ppm in a paper on burning chipped tires as a hog fuel supplement. Hunter reported values of 138 to 395 ppm  $\text{NO}_x$  from two boilers burning a mixture of hog fuel and coal.

The earliest reference discussed to date on  $\text{NO}_x$  emissions from any type of wood combustion is by the Stanford Research Institute (1950) which gave a value of 1.3 pounds of nitrogen oxide (as  $\text{NO}_2$ ) per ton of wood waste burned in a silo incinerator.

In preparation for this research investigation, the author tested three hog fuel fired boilers during the summer of 1976 (Kester 1976) using grab sampling techniques and reported concentrations of nitrogen oxides ranging from 40 to 75 ppm. The boilers tested burned hog fuel of about 10 percent and 50 percent moisture content.

The lower NO<sub>x</sub> concentration was measured in the boiler burning the wetter fuel, and the higher concentrations were measured with the drier fuel burned. It was not determined if the difference in concentrations was associated with fuel moisture content or other variables such as combustion temperature or fuel feed rate.

C. NITROGEN OXIDES

Nitrogen oxides are a primary pollutant that react photochemically to form secondary pollutants. EPA is focusing ever-increasing attention at the problem of secondary reaction oxidant contamination. The author has recently been contacted by EPA regarding developing emission standards for new hog fuel boilers. EPA is looking into this area and is trying to obtain data on current or previous work on the subject. The EPA ambient standard is 0.05 ppm NO<sub>2</sub> on an annual average. California now has an ambient 1-hour 0.25 ppm nitrogen oxides standard and EPA is considering adopting a short-term 3-hour ambient nitrogen oxides standard.

EPA is also now looking into the area of fine atmospheric particulates. SO<sub>2</sub> and NO<sub>x</sub> have been identified as important contributors to the fine particulate

load through atmospheric gas to particle reactions resulting in sulfates and nitrates (Waggoner, 1979).

The major component of worldwide atmospheric nitrogen oxides is biologically produced nitric oxide (NO). Natural sources produce about  $50 \times 10^7$  tons of NO per year; manmade sources, primarily fuel combustion, emit  $5 \times 10^7$  tons per year of  $\text{NO}_x$ , which include NO and  $\text{NO}_2$ .

Of the various oxides of nitrogen, the most important as air pollutants are NO and  $\text{NO}_2$ . Other known oxides of nitrogen include nitrous oxide ( $\text{N}_2\text{O}$ ), nitrogen sesquioxide ( $\text{N}_2\text{O}_3$ ), nitrogen tetroxide ( $\text{N}_2\text{O}_4$ ), and nitrogen pentoxide ( $\text{N}_2\text{O}_5$ ); however, only  $\text{N}_2\text{O}$  is present in the atmosphere in appreciable concentrations.

Nitric oxide and a comparatively small amount of  $\text{NO}_2$  are formed under high-temperature conditions such as those that accompany the burning of fossil fuels. They are emitted to the atmosphere from automobile exhausts, furnace stacks, incinerators, and vents from certain industrial processes. Although vast quantities are produced by natural biological reactions, the resultant concentrations of atmospheric  $\text{NO}_x$  are low because these reactions take place over wide areas.

Most of the  $\text{NO}_x$  produced by man is in the form of NO, which is subsequently oxidized in the atmosphere to the more toxic and irritant  $\text{NO}_2$ . Normally, at low NO concentrations of 1 ppm or less, the direct reaction with oxygen of the air proceeds slowly. The oxidation rate of NO to  $\text{NO}_2$  is increased by photochemical processes involving reactive hydrocarbons.

The following describes the properties of the major oxides of nitrogen:

#### Nitric Oxide (NO)

Nitric oxide is a colorless, odorless gas, slightly soluble in water. It is the primary product formed during high-temperature combustion processes when atmospheric oxygen and nitrogen combine according to the reactions described in Section III. When rapid cooling follows combustion, time is inadequate for equilibrium to develop, and NO persists in the flame products.

#### Nitrogen Dioxide ( $\text{NO}_2$ )

Nitrogen dioxide is a reddish-orange-brown gas with a characteristic pungent odor. The threshold for odor

perception of  $\text{NO}_2$  is about 0.12 ppm. Though its boiling point is 70 degrees F, the low partial pressure of  $\text{NO}_2$  in the atmosphere restricts it to the gas phase at usual atmospheric temperatures.  $\text{NO}_2$  is corrosive and highly oxidizing and may be physiologically irritating and toxic. Although  $\text{NO}_2$  reacts with water to form nitric acid and either nitrous acid or  $\text{NO}$ , the reaction of  $\text{NO}_2$  with  $\text{H}_2\text{O}$  has not been shown to have any significance in polluted air (Leighton, 1961).

Although  $\text{NO}_2$  exists in equilibrium with its dimer, nitrogen tetroxide ( $\text{N}_2\text{O}_4$ ), the fraction present in dimer form is negligible at atmospheric concentrations of  $\text{NO}_2$ .

#### Nitrous Oxide ( $\text{N}_2\text{O}$ )

Nitrous oxide is a colorless and odorless gas used as an anesthetic and is familiarly known as "laughing gas".  $\text{N}_2\text{O}$  is present in the atmosphere but is chemically inert at usual atmospheric temperatures and is, therefore, not considered an air pollutant.  $\text{N}_2\text{O}$  may be a minor product of the atmospheric interactions of  $\text{NO}_2$  in the presence of hydrocarbons and sunlight. Nitrous

oxide is also a possible intermediate in thermal NO formation.

#### Nitrogen Sesquioxide ( $N_2O_3$ )

Nitrogen sesquioxide may be involved as an intermediate in atmospheric reactions. It is the anhydride of nitrous acid and produces this acid when dissolved in water. In the laboratory, it can be obtained as a blue liquid by cooling a mixture of nitric oxide and nitrogen dioxide.

#### Nitrogen Tetroxide ( $N_2O_4$ )

Nitrogen tetroxide is the colorless dimer of nitrogen dioxide. Its formation is favored by high  $NO_2$  concentrations. At the dilute  $NO_2$  concentrations found in ambient atmospheres, however,  $N_2O_4$  cannot be present in more than trace amounts and therefore is probably of minor importance in atmospheric reactions.

#### Nitrogen Pentoxide ( $N_2O_5$ )

Like  $N_2O_3$ ,  $N_2O_5$  may be an intermediate in atmospheric reactions.  $N_2O_5$  is the anhydride of nitric acid and

presumably reacts with atmospheric moisture to form nitric acid.

### Nitrogen Trioxide ( $\text{NO}_3$ )

Nitrogen trioxide is thought to be an important intermediate in atmospheric reactions, but equilibrium and reaction kinetics factors do not favor accumulation of significant (or measurable) levels. Both  $\text{NO}_3$  and  $\text{NO}_2$  have been suggested as intermediates during the formation of  $\text{NO}_2$  when NO reacts with  $\text{O}_2$ .

## D. EFFECTS OF NITROGEN OXIDES

### Visibility

Visibility reduction is one of the most common manifestations of urban air pollution. It is caused by the scattering and absorption of light by particles and gases in the atmosphere and depends on the concentration and properties of the gases and particles present.

$\text{NO}_2$  is intensely colored and absorbs light over the entire visible spectrum, but primarily in the shorter wavelengths, violet, blue, and green. In the atmosphere it can reduce the brightness and contrast of

distant objects and causes the horizon sky and white objects to appear pale yellow to reddish-brown. However, the primary cause of the characteristic brown haze of typical urban air pollution is light scattering by aerosol particles, not  $\text{NO}_2$ .

The additional presence of particulate matter tends to mask the coloration effect of  $\text{NO}_2$ , but the two combined markedly reduce the visibility, contrast, and brightness of distant objects. Particulate matter and aerosols are present in the atmosphere as primary contaminants from urban sources such as industrial combustion and vehicular transportation and from natural sources such as sea salt, dust, and fog. They are also formed through photochemical reactions and are considered to be the major cause of the reduced visibility associated with photochemical smog.

The photochemical reactions involves  $\text{NO}_x$  and hydrocarbons under the influence of sunlight. The ultraviolet sunlight has the energy to break an O atom from a  $\text{NO}_2$  molecule. The O then combines with  $\text{O}_2$  forming ozone, which can oxidize the hydrocarbons present, resulting in the formation of visibility reducing aerosols. Light scattering associated with the presence of aerosols is the primary cause of visibility

reduction in photochemical smog. Absorption of light by  $\text{NO}_2$  makes a minor contribution (Charlson, 1972).

### Concentrations

Air quality standards limiting amounts of  $\text{NO}_x$  are primarily aimed at limiting the amount of photochemical smog, as measured as ozone, produced. Both NO and  $\text{NO}_2$  concentrations display distinct diurnal variations dependent on both the intensity of the solar ultra-violet energy and the amount of atmospheric mixing. In many sampling areas, these variations are also associated with the traffic patterns.

The effect of meteorological parameters on NO and  $\text{NO}_2$  concentrations has been reasonably well documented. As might be expected, periods of stagnation and high traffic volume in urban areas have resulted in high peak levels of  $\text{NO}_x$ , as much as 3 ppm as a peak in Los Angeles for example.

Continuous measurement has indicated that peak values of NO above 1 ppm are widespread, but  $\text{NO}_2$  concentrations have rarely been measured at this level. Peak concentrations of  $\text{NO}_2$  in urban areas rarely exceed 0.5 ppm.

### Vegetation

In terms of direct effects on growing vegetation, the major sources of high levels of nitrogen dioxide are localized, accidental releases which cause relatively short periods of exposure. These high concentration exposures produce necrotic lesions and excessive defoliation. Such lesions are usually characteristic for each plant, but their nonspecific character in relation to other toxicants renders the symptoms of little value in diagnosing NO<sub>2</sub> damage in the field. There is, however, no direct relationship between time of exposure and concentration, except within very narrow ranges. The concentration of nitrogen dioxide influences the extent of injury more than the duration of exposure.

### Materials

Significant effects of NO<sub>x</sub> have been observed and studied on three classes of materials: textile dyes and additives, natural and synthetic textile fibers, and metals. The most pronounced problem is associated with textile dyes and additives. Fading and loss of color are the primary problem in textile fibers, while

particulate nitrates and humidities above 50 percent lead to stress corrosion in metals.

### Health

Both of the prominent oxides of nitrogen present in ambient air are potential health hazards. At ambient concentrations, NO presents no direct threat to general health; however, NO<sub>2</sub> does. Nitric oxide is not an irritant and is not considered to have adverse health effects at concentrations found in the atmosphere. Its greatest toxic potential at ambient concentrations is related to its tendency to undergo oxidation to NO<sub>2</sub>. NO<sub>2</sub> exerts its primary toxic effect on the lungs. High concentrations, greater than 100 ppm, are lethal to most animal species; 90 percent of the deaths are caused by pulmonary edema.

Since certain pathological changes seen in animals after experimental NO<sub>2</sub> exposure are similar to changes that occur in man, it is suggested that long-term low-level exposures to NO<sub>2</sub> may play a significant role in the development of chronic lung disease. However, the small amount of information

available concerning the toxicological effects of the oxides of nitrogen in man pertain to levels higher than those found in ambient air (Barth, 1970)

### III. FORMATION OF NITROGEN OXIDES IN FLAMES

#### WOOD COMBUSTION

The proximate and ultimate analysis of the Douglas fir bark used in this study is given table 3 below (Tuttle, 1978).

Table 3. ANALYSIS OF DOUGLAS FIR BARK

	<u>Percent by Weight (dry basis)</u>
<u>Proximate Analysis</u>	
Volatile matter	71.0
Fixed Carbon	28.0
Ash	1.0
<u>Ultimate Analysis</u>	
Hydrogen	6.1
Carbon	54.1
Nitrogen	0.17
Oxygen	38.8
Sulfur	0.0
Ash	1.0

Wood normally contains some moisture. Kiln dried wood contains about 8 percent; air dried wood contains 15 to 25 percent; green wood contains approximately 30 to 60 percent; and wet bark may contain up to 75 percent water by weight. These values and all others expressed in this work are on a "wet" basis, that is, pounds of moisture per pound of wet wood. Therefore, a piece of wood that was half water

and half wood would have a moisture content of 50 percent. Typical caloric values are 8,000 to 9,000 Btu per pound for dry wood and 9,000 to 9,300 Btu per pound for dry bark. The Douglas fir bark used in this experiment has a higher heating value of 9,290 Btu per bone dry pound.

The pyrolytic decomposition and combustion of wood are a series of complex and not universally agreed upon physical-chemical reactions that occur primarily in the gaseous phase. However, in a simplistic way, the combustion can be described in four basic steps (Corder, 1965; Junge, 1975; Kreisinger, 1939). As fresh wood enters the furnace, moisture is first driven off in an endothermic reaction. Secondly, the combustible volatiles are then distilled from the wood. This is also an endothermic reaction requiring heat, but these volatiles are then available to burn in an exothermic reaction providing heat to both the boiler and the fuel pile in the third step. Finally, after the moisture and volatiles have been driven from the wood, the fixed carbon remains. In this fourth step, this carbon is also oxidized in an exothermic reaction, similar to burning charcoal briquets in a home barbecue.

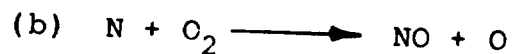
Approximately 70 to 80 percent of the heating value of the wood is contained in the distillates and the remaining 20 to 30 percent in the fixed carbon. Since the predominant

elements in wood are carbon, hydrogen, and oxygen, the complete combustion products are almost entirely carbon dioxide and water.

### NITROGEN OXIDE FORMATION

The reaction of atmospheric oxygen and nitrogen to form NO is associated principally with the high temperature, fuel lean conditions in industrial boiler combustion chambers. Normal operation of a typical hog fuel boiler is in this fuel lean mode.

In most high temperature combustion processes, the formation of NO from the dissociation of atmospheric nitrogen is considered to be primarily due to the Zeldovich (1946) mechanism, equations (a) and (b).



NO<sub>2</sub> is formed by the further oxidation of NO, mostly after the combustion gases have left the furnace; this process is known as the fixation of atmospheric nitrogen. The NO<sub>x</sub> produced via the mechanism is termed "thermal NO<sub>x</sub>" due to its exponential temperature dependence.

At lower combustion temperatures, the Zeldovich mechanism predicts low  $\text{NO}_x$  values compared to actual data. Mechanisms which would account for such discrepancies (Malte, 1977) are ones in which  $\text{N}_2\text{O}$  forms first by the reaction  $\text{N}_2 + \text{O} + \text{M} \rightarrow \text{N}_2\text{O} + \text{M}$  and acts as a precursor to NO formation via the reaction  $\text{N}_2\text{O} + \text{O} \rightarrow 2\text{NO}$ . Another mechanism which may be active at moderate combustion temperatures is the "prompt NO" mechanisms (Fenimore, 1971). The initiating reaction for prompt NO generation is thought to be  $\text{N}_2 + \text{CH} \rightarrow \text{NCH} + \text{N}$ . Further oxidation of the products of this reaction yields NO.

Additionally, at moderate combustion temperatures, the Zeldovich mechanism may still contribute to  $\text{NO}_x$  formation. This is because combustion in an industrial boiler furnace is turbulent. Although the first Zeldovich reaction has an extremely high activation energy and the average gas temperatures in an industrial boiler are low, there will be areas of higher temperatures due to the turbulent nature of the combustion. This will extend the effect of the Zeldovich mechanism to lower average furnace temperatures (Malte, 1978).

Higher local temperatures may also be reached in the fixed carbon (char) bed on the grates of the furnace. Temperatures greatly in excess of the gas temperature have been

measured in the char bed during other experiments in this combustor. (Tuttle 1979).

An assumption in this study was that  $\text{NO}_x$  mechanisms of all types present are independent and additive. It is because of the complex nature of  $\text{NO}_x$  formation, the complex nature of wood combustion, and the complex nature of industrial hog fuel boiler operation that an experimental approach, rather than an attempt at a rigorous combustion reaction investigation, was undertaken for this study.

Engleman (1973) discusses the difficulty of a rigorous approach to investigating  $\text{NO}_x$  emissions from complex fuels in which he shows that actual  $\text{NO}_x$  formation follows theory well for a hydrogen/air flame, nearly as well for a carbon monoxide/air flame, and not well at all for a propane/air flame. Hog fuel/air is expected to be more complex than any of the above. He states:

"Reactions which have little significance in the over-all combustion scheme may be of importance in  $\text{NO}$  formation under specific conditions. The evaluation of the reactions significant for the characterization of gross combustion phenomena is a formidable task by itself. The numerous additional reactions, which may

be significant under specific conditions for the formation of parts per million of nitric oxide, present even more difficult problems."

As the kinetic mechanisms become more complex, as they would in a bark/air combustion scheme, critical evaluation of the component reactions becomes increasingly important. The coupling of kinetic modeling with sophisticated fluid mechanics and heat transfer modeling would be ultimately required for application to  $\text{NO}_x$  formation in actual combustion systems. Such a study is beyond the scope of this work.

Another source of  $\text{NO}_x$  is oxidation of nitrogen bound in the fuel. Wide ranges of fuel nitrogen to  $\text{NO}_x$  conversion have been reported for fossil fuels (Malte, 1978), but there have been no reported studies of fuel nitrogen conversion in hog fuel fired boilers. In this study, however, the fuel nitrogen content was quite low and constant at 0.17 percent. The variability of NO concentration with fuel nitrogen was not considered in this experiment. It has been shown by Fenimore (1971) and others that fuel bound nitrogen in various fossil fuels is converted to NO. It is assumed, but has not been shown, that the same phenomenon occurs during wood combustion. Because of the low concentration of nitrogen in bole wood and bark,

usually reported as 0.1 percent, most typical hog fuel boilers probably have a small portion of their total nitrogen oxide emissions contributed by the fuel nitrogen portion. However, there is potential for this fuel nitrogen content to be greatly increased in hog fuel combustion.

Presently there is interest being shown by wood products industries and power-generating utilities in investigating the setting aside of large tracts of land to continuously grow and harvest whole trees for use as fuel. This is the so-called Biomass concept, in which the area dedicated to growing trees is sufficient to continuously supply the necessary hog fuel for the subject plant or industry. Seattle City Light is presently studying such a project in King County using alder trees.

Because of the volume requirements of storing a long-term supply of hog fuel and the logistics problems of logging during the winter (when the leaves would be off the trees, reducing the total nitrogen content) in some parts of the country, it will be necessary to harvest the trees on a year-round basis. The nitrogen in trees is primarily concentrated in the living cells of the trees, especially the leaves and small branches (Dice, 1970). Therefore, when these whole trees are cut for fuel, the nitrogen content of the whole-tree fuel will be considerably higher than

that of conventional hog fuel. These higher levels of fuel nitrogen may lead to higher nitrogen oxide emissions.

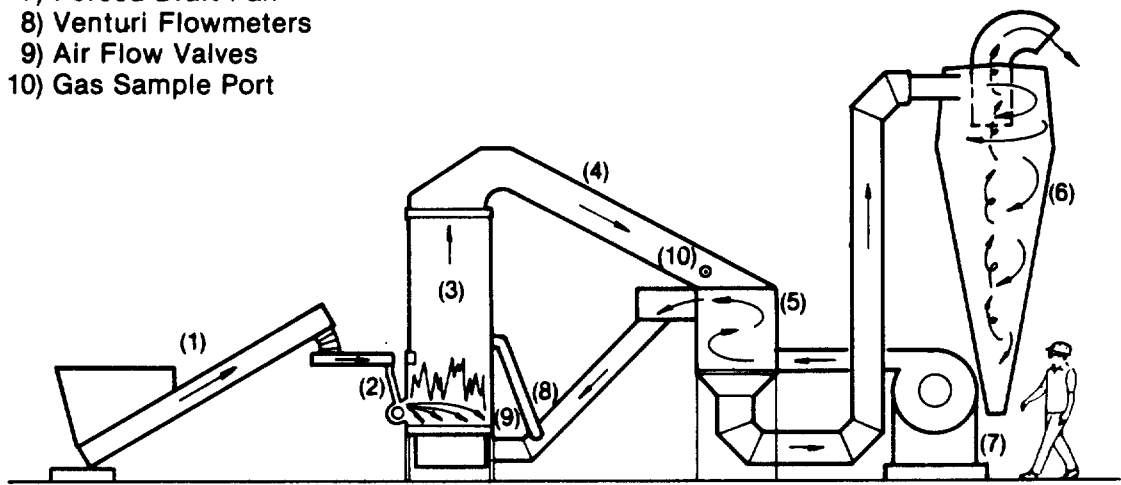
#### IV. EXPERIMENTAL TEST FACILITY

##### A. PILOT BOILER

Data collected during the course of the experiments were obtained from the operation of a pilot scale spreader stoker boiler and combustion test facility. The boiler and test facility, shown in figure 3, is described as follows:

1. A dual screw conveyor system to control and meter the fuel flow.
2. A mechanical flinger roll (spreader stoker) to distribute fuel uniformly across the grate.
3. A water wall lined combustion chamber with a fixed, pinhole grate.
4. A fin tube, forced air cooled, jacketed duct to carry combustion exhaust gases from the combustion zone to an air preheater.
5. A plate type, counter flow, triple pass air preheater.

- 1) Conveyor System
- 2) Spreader Stoker
- 3) Combustion Chamber
- 4) Air Cooled Duct
- 5) Heat Exchanger
- 6) Cyclone
- 7) Forced Draft Fan
- 8) Venturi Flowmeters
- 9) Air Flow Valves
- 10) Gas Sample Port



**Wood Combustion Test Facility**

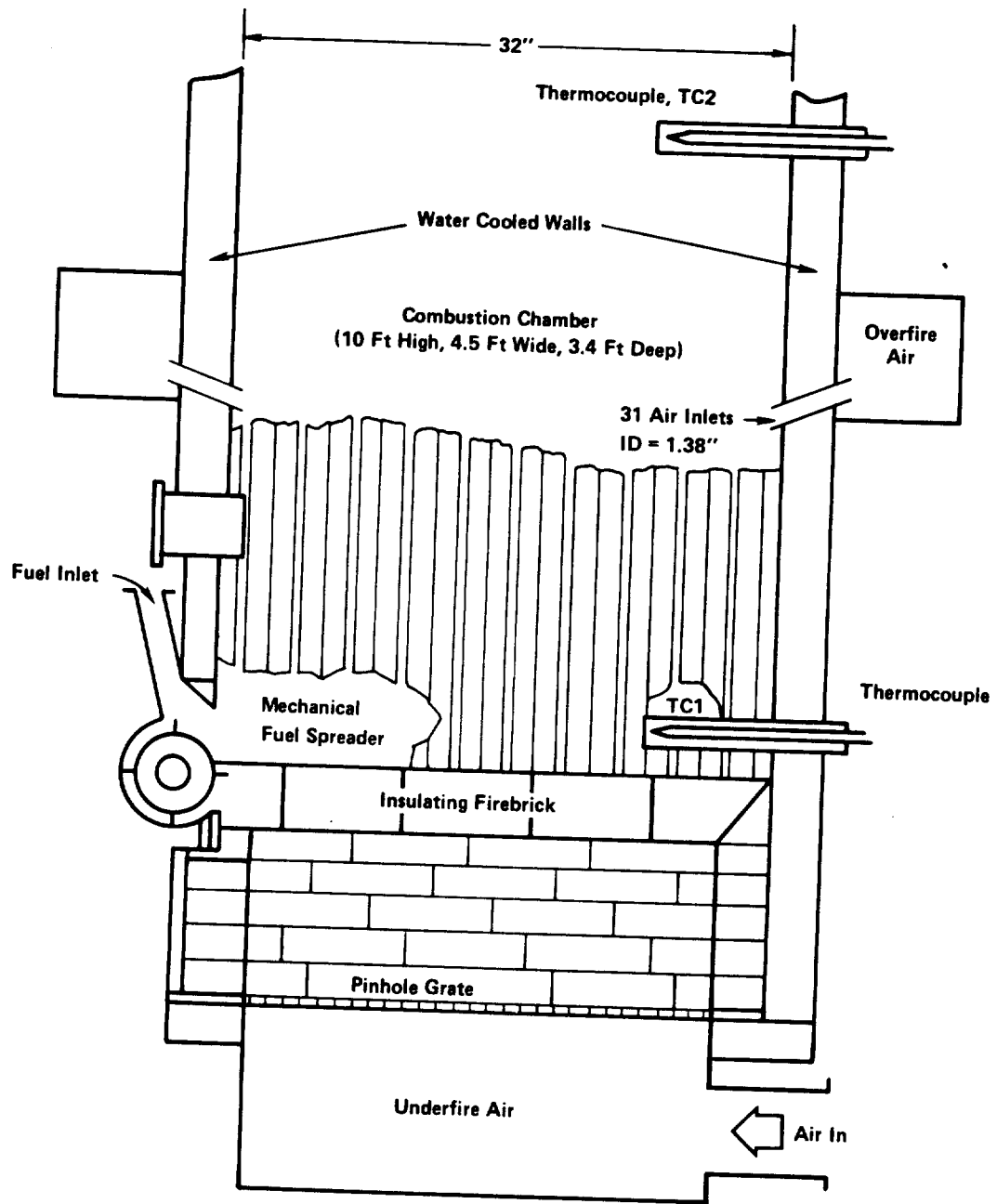
**FIGURE 3**

6. A cyclone separator downstream from the air pre-heater to remove entrained particulate matter from the exhaust gas stream.
7. A forced draft fan to provide combustion air to the test facility.
8. A system of venturi orifice flowmeters to measure the amount of air distributed above and below the grate.
9. A system of gate valves to control the flow of air above and below the grate.
10. Instrumentation to measure independent and dependent variables of the test facility.

The boiler was designed to combust fuel samples with heat release rates of up to  $10 \times 10^6$  Btu's per hour. Typical tests are carried out with heat release rates in the range of  $6 \times 10^6$  Btu's per hour. The area of the grate is 10 square feet. The combustion zone extends approximately 11 feet above the grate. Steam is generated at atmospheric pressure with natural recirculation in the water walls (Junge, 1977).

Typical tests were carried out in the 4.2 and 7.0 million Btu/hr range. This resulted in heat release rates of 23,300 to 38,800 Btu per cubic foot per hour. The pilot plant was operated in the steady state in essentially the same manner as full-scale industrial hog fuel boilers.

Figure 4 is a view of the combustion chamber taken in vertical cross section. A rotating, mechanical spreader distributes the fuel evenly across the grate. The water wall construction begins above the spreader stoker and continues up to the top of the furnace. The ash removal doors are below the spreader stoker. Ten pounds of charcoal can be inserted through the ash door for light off. Under fire air passes up through the pinholes in the grate. Over fire air enters through 1½-inch diameter nozzles, between the nonpressurized water tubes, 42 inches above the grate. The over fire air ports are directed slightly downward, approximately 12 degrees. The flow rates of air, both under and over fire are controlled by dampers in the supply lines at the entrance to the plenum chambers. Combustion chamber temperatures are indicated by single radiation-shielded thermocouples inserted between water tubes at levels above the grate of 20 and 63 inches. Above the combustion chamber,



Combustion Chamber, Side View

FIGURE 4

an air cooled duct leads approximately 14 feet to the plate type, counter flow, air preheater.

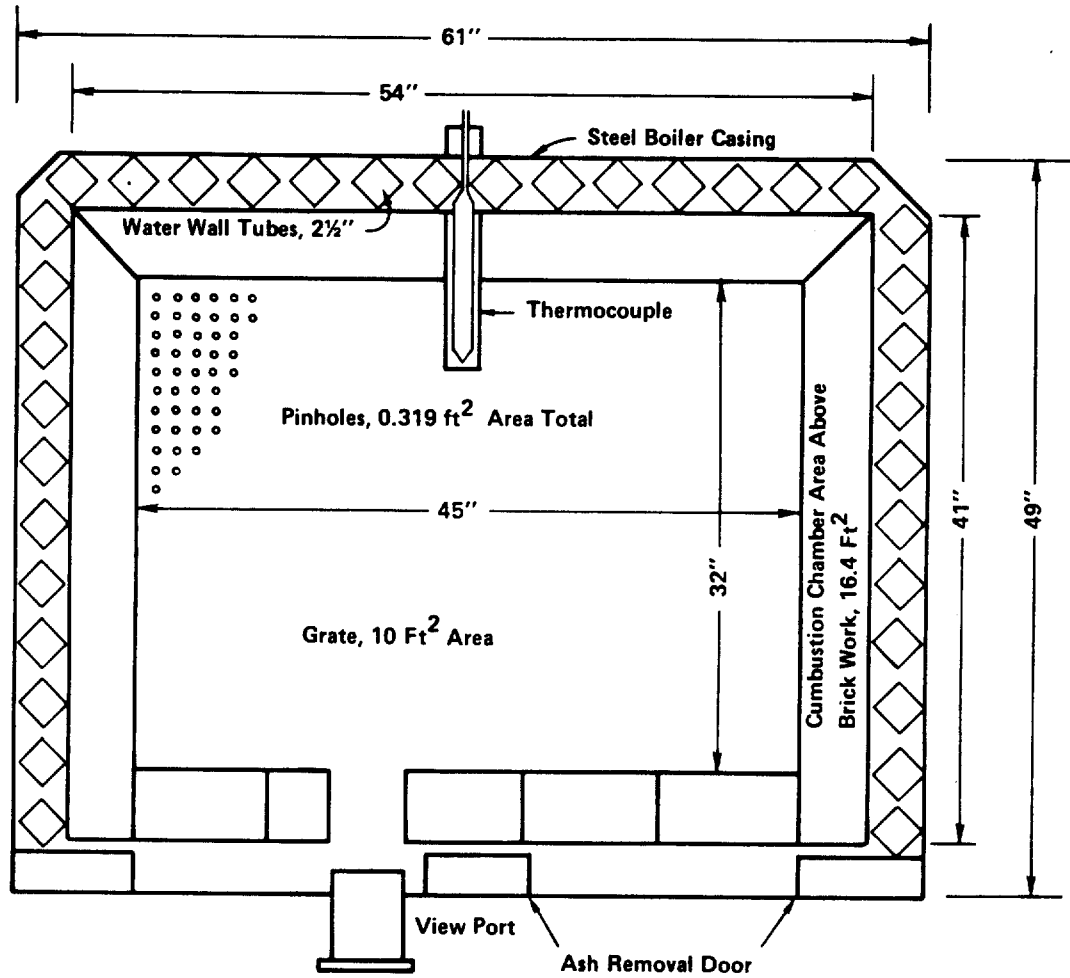
Figure 5 shows the lower part of the combustion chamber in horizontal cross section. The 16.4 square feet of combustion chamber area are reduced to 10.0 square feet at the grate by a 4½-inch-wide, 13-inch-high fire brick lining. The grate has quarter-inch pinholes on 1.25-inch centers in quarter-inch stainless steel plate. The arrangements of the square steel water wall tubes and steel outer casing are shown in figures 4 and 5 with dimensions.

## B. MEASUREMENT METHODS AND PROCEDURES

The following section describes the measurement methods and procedures for the study. A more detailed description of those measurements and procedures that relate to a major potential for error in the data is presented in Section VI D, "Experimental Errors."

### 1. Nitric Oxide (NO)

The nitric oxide was measured with a Lear Siegler model SM 1000 ultraviolet analyzer. This



Lower Combustion Chamber, Cross Section

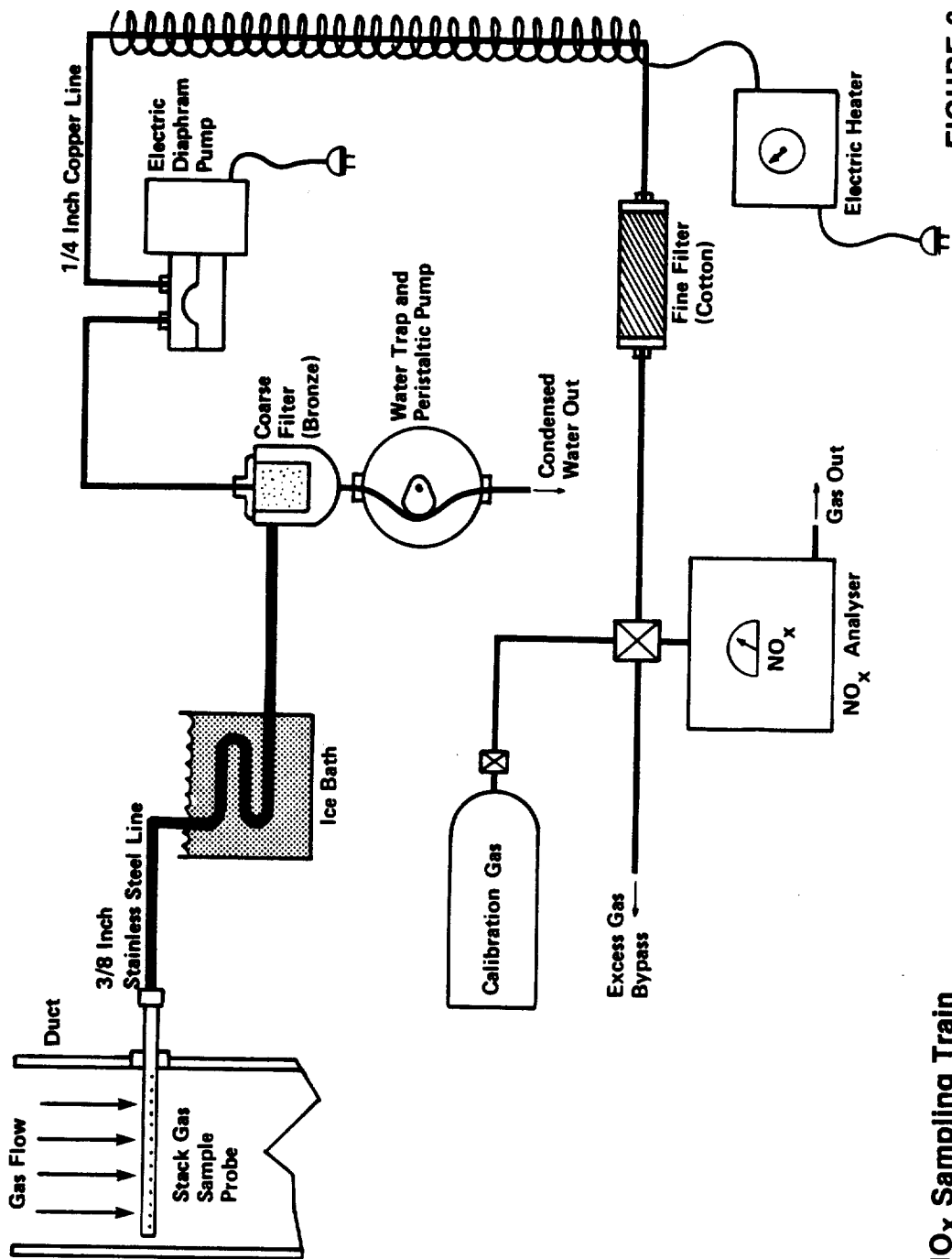
FIGURE 5

instrument was calibrated using a 50 ppm NO sealed internal span cell and 100 ppm NO laboratory span gas.

The gas sample was withdrawn at a flow rate of 1 cfm upstream of the air preheater through the gas sample port. The NO sampling train, shown schematically in figure 6, consisted of a stainless steel probe, followed by an ice bath, a coarse filter (sintered bronze), a water trap equipped with a peristaltic water pump to drain the trap, a diaphragm gas pump, a heated copper tube (heat tape wrapped to raise the gas temperature to 80 degrees to 100 degrees F), a fine filter (cotton in a 2-inch-diameter glass tube 12 inches in length), and on to the NO analyzer. The gas sampling line was 3/8-inch-diameter stainless steel tubing up to the water trap and 1/4-inch copper tubing from the water trap to the NO analyzer. There was a bypass for the gas sample flow rate in excess of the 400cc/min needed by the NO analyzer.

## 2. Fuel Feed Rate

The fuel feed rate was measured by weighing the bark fuel delivered by the screw conveyers for a



measured period of time. The fuel feed rate was measured at various speeds of each screw conveyer in order to calibrate the variac and ammeters on the variable speed electric motor. Fuel samples were dried and moisture contents measured so that fuel feed rates in bone dry pounds per hour were known. Fuel feed rates varied from 450 to 750 pounds per hour (bone dry).

### 3. Excess Air

The total flow (under fire plus over fire) to the combustion chamber was controlled using damper valves with venturi flowmeters and Magnahelic pressure differential gauges.

The desired amount of combustion air flow was calculated for each given percent excess air by stoichiometric combustion calculations. The combustion air flow rate varied from about 1,100 to 2,600 acfm at about 280 degrees F. The combustion air flow rate was set to operate at 30, 50, and 80 percent excess air for the various fuel rates.

#### 4. Gas Temperature

The combustion gas temperature was measured with a shielded platinum-rhodium thermocouple located 63 inches above the grate.

#### 5. Fuel Size

The fuel was prescreened into two size fractions. The coarse fuel ranged from 0.3 to 1.3 inches diameter; the fine fuel was 0.3 inch and under.

#### 6. Fuel Moisture Content

The fuel moisture content was measured by weighing a fuel sample before and after drying to constant weight at 219 degrees F.

#### 7. Under Fire Air Flow Rate

The under fire air flow rate was measured using a venturi flowmeter and differential pressure gauge (Magna-helic). The desired under fire air flow rate was reported in lb air/lb bone dry fuel and ranged from 1.5 to 6.5 pounds of air per pound bone dry fuel. With the percentages of

carbon, hydrogen, and oxygen given in table 1 for the Douglas fir bark fuel, stoichiometric combustion calculations show that 6.7 pounds of air are required for complete combustion of the fuel.

## V. EXPERIMENTAL RESULTS

### A. NITROGEN OXIDE DATA BASE

A data base was developed showing the effects of the fuel feed rate, combustion temperature, excess air ratio, fuel size, fuel moisture content, and undergrate air flow upon  $\text{NO}_x$  concentrations. This data base and the relationships are shown graphically in figures 7 through 13.

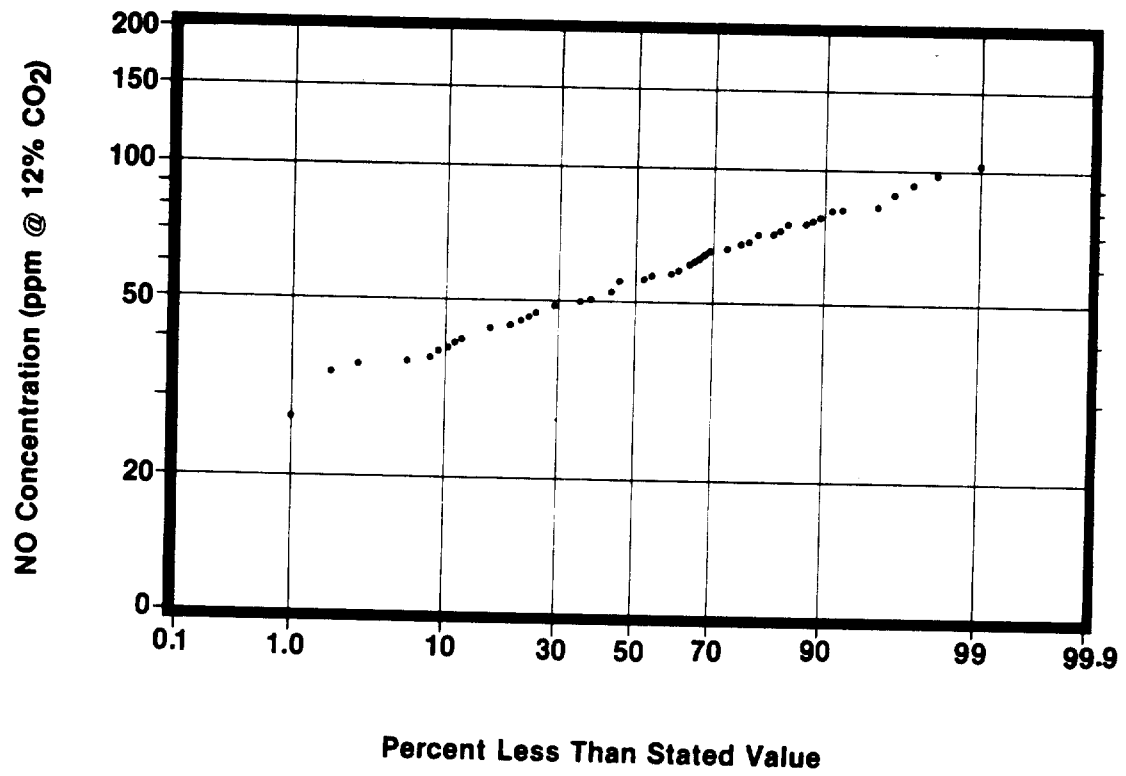
#### 1. NO Concentrations

The NO concentrations were recorded at various fuel feed rates, percent excess air, various fuel feed rates, percent excess air, under fire air flow rates, fuel size, and moisture content.

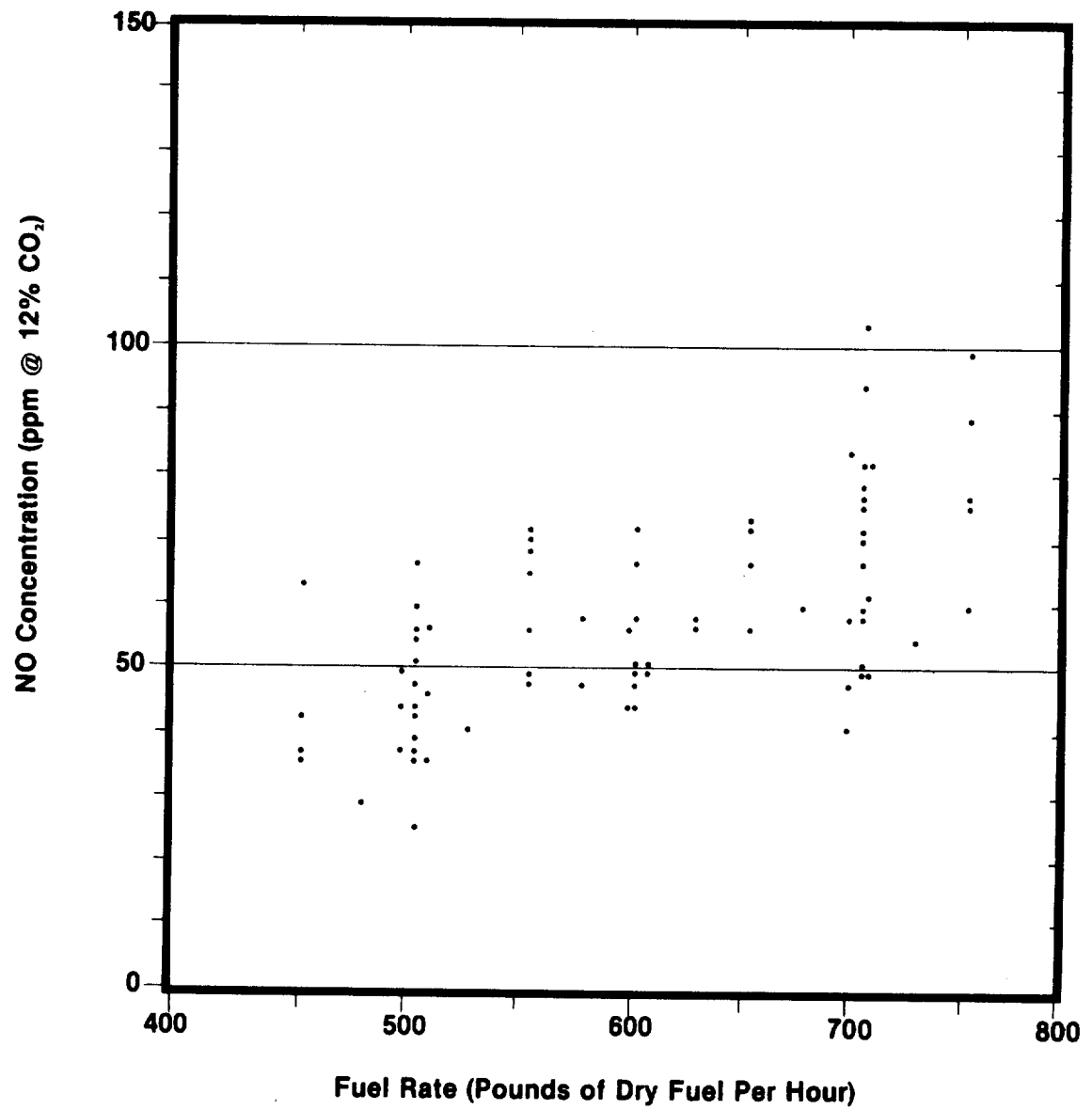
The 97 NO values, corrected to 12 percent  $\text{CO}_2$  and on a dry basis, range from 25 to 103 ppm with an average of 57 ppm, as shown in figure 7.

#### 2. Effect of Fuel Feed Rate (RATE)

The effect of fuel feed rate on the NO concentration is shown in figure 8. The NO concentration



Percent Less Than Stated Value Per NO Concentration **FIGURE 7**



Fuel Rate Per NO Concentration

FIGURE 8

increases with increasing fuel feed rate. The coefficient of correlation is 0.645.

### 3. Effect of Combustion Temperature (TEMP)

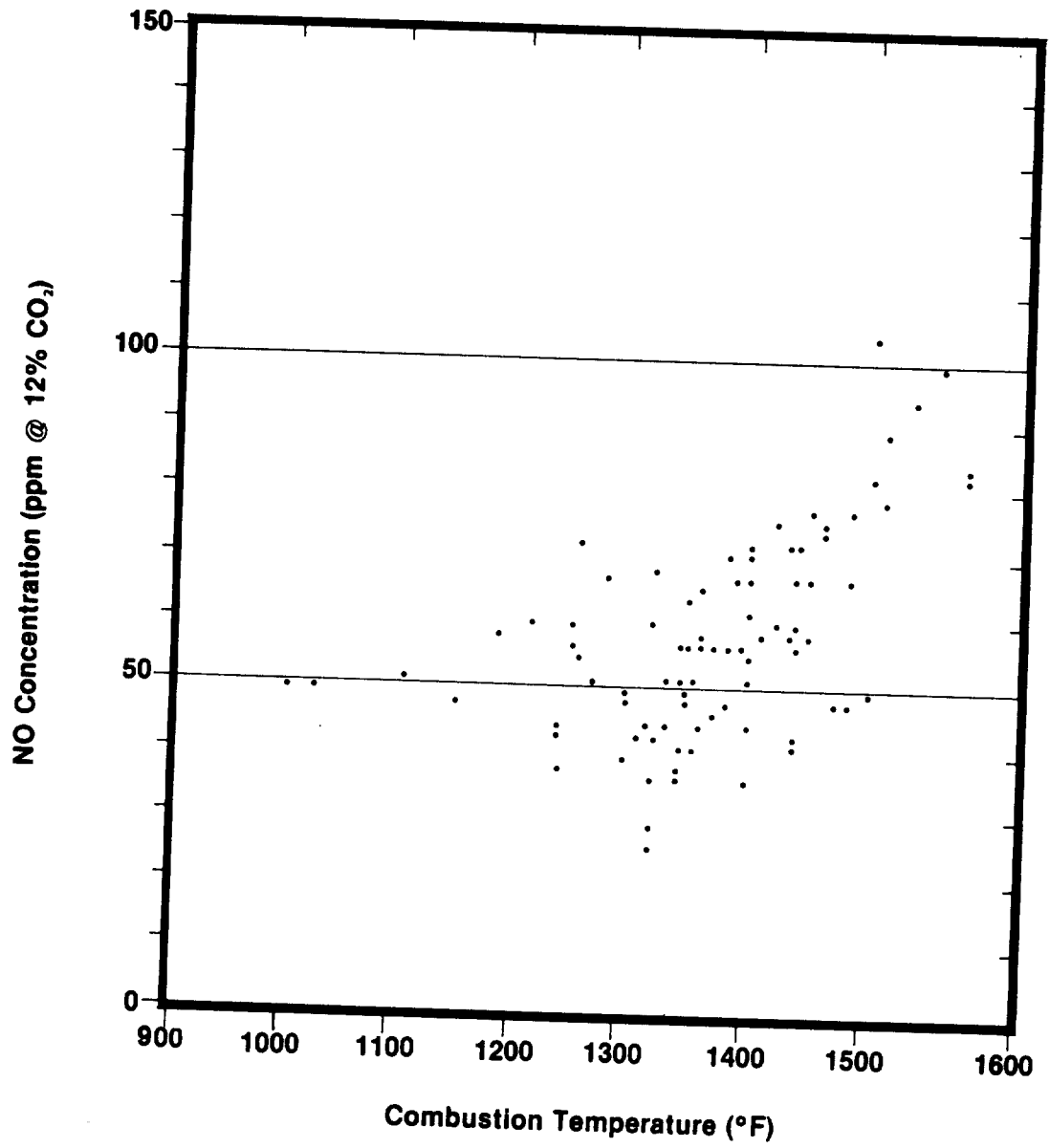
The effect of combustion temperature on the NO concentration is shown in figure 9. The NO concentration increases with increasing combustion temperature. The coefficient of correlation is 0.549.

### 4. Effect of Excess Air (XAIR)

The effect of excess air on the NO concentration is shown in figure 10. The NO concentration increases slightly with increasing excess air. The coefficient of correlation is 0.340.

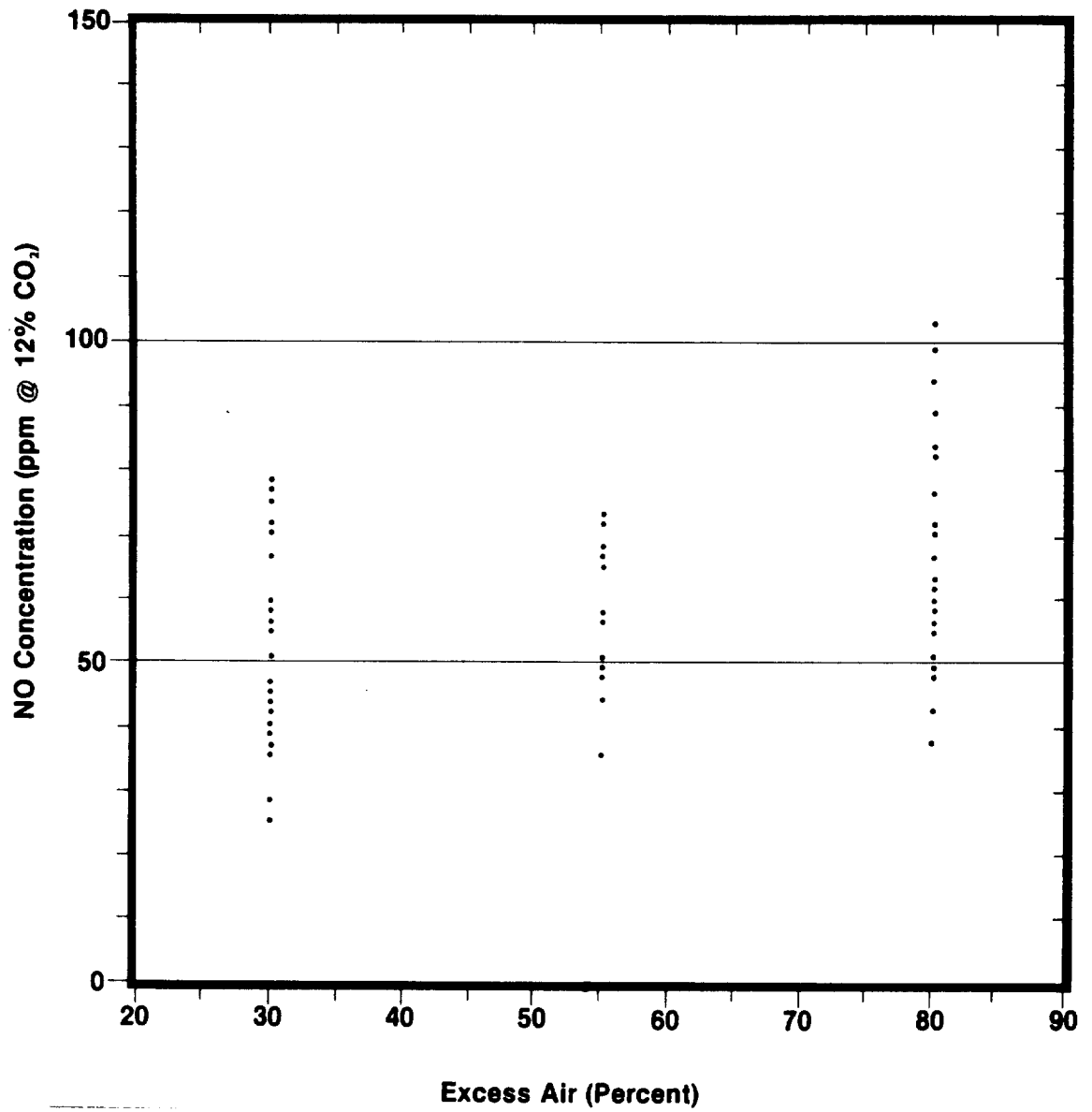
### 5. Effect of Fuel Size (SIZE)

The effect of fuel size on the NO concentration is shown in figure 11. There appears to be a slight decrease in the NO concentration with increasing fuel size. The coefficient of correlation is -0.199.



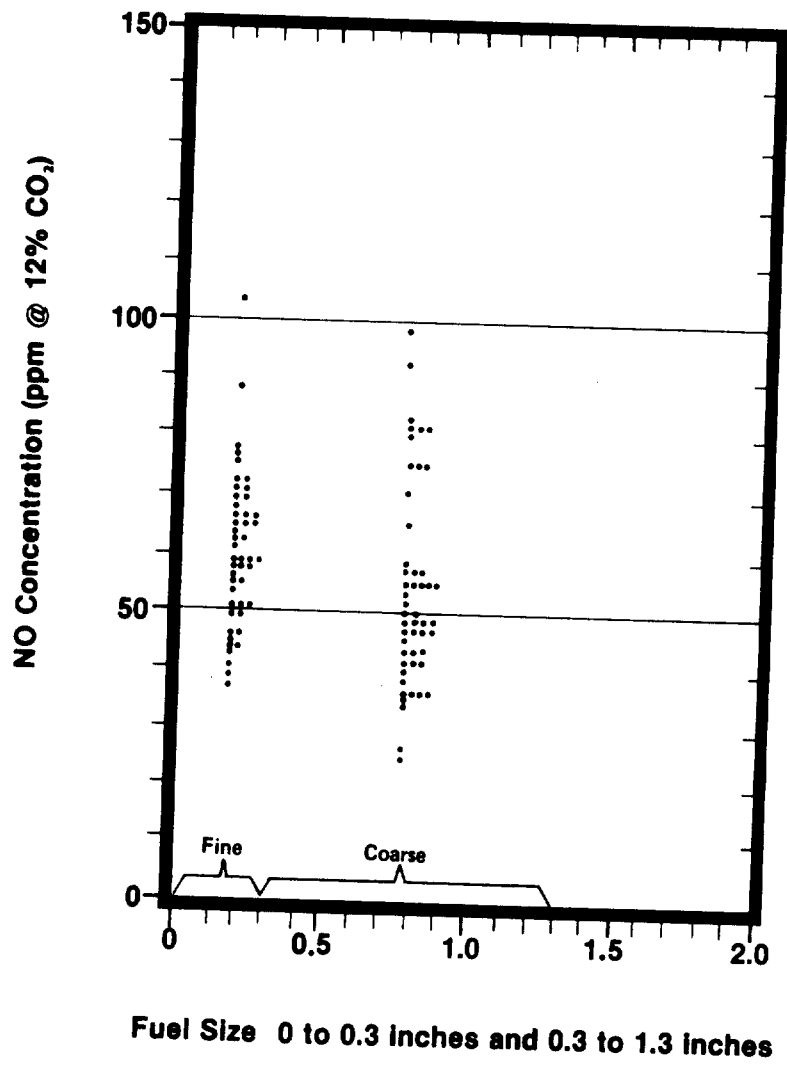
Combustion Temperature Per NO Concentration

FIGURE 9



Excess Air Per NO Concentration

FIGURE 10



Fuel Size Per NO Concentration

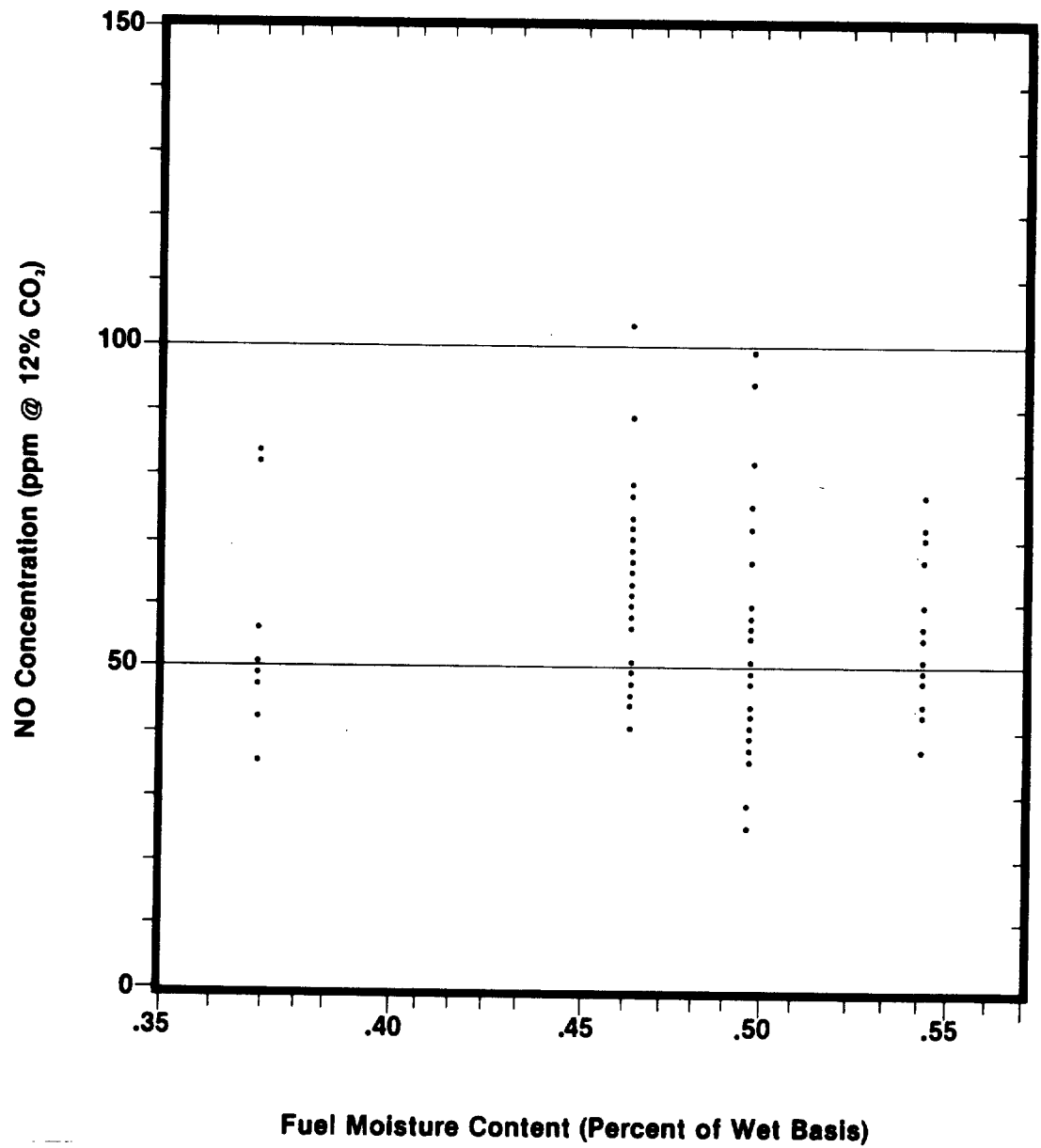
FIGURE 11

6. Effect of Fuel Moisture Content ( $H_2O$ )

The effect of fuel moisture content on the NO concentration is shown in figure 12. There appears to be a very slight decrease in NO concentration with an increase in fuel moisture content. The coefficient of correlation is -0.0656.

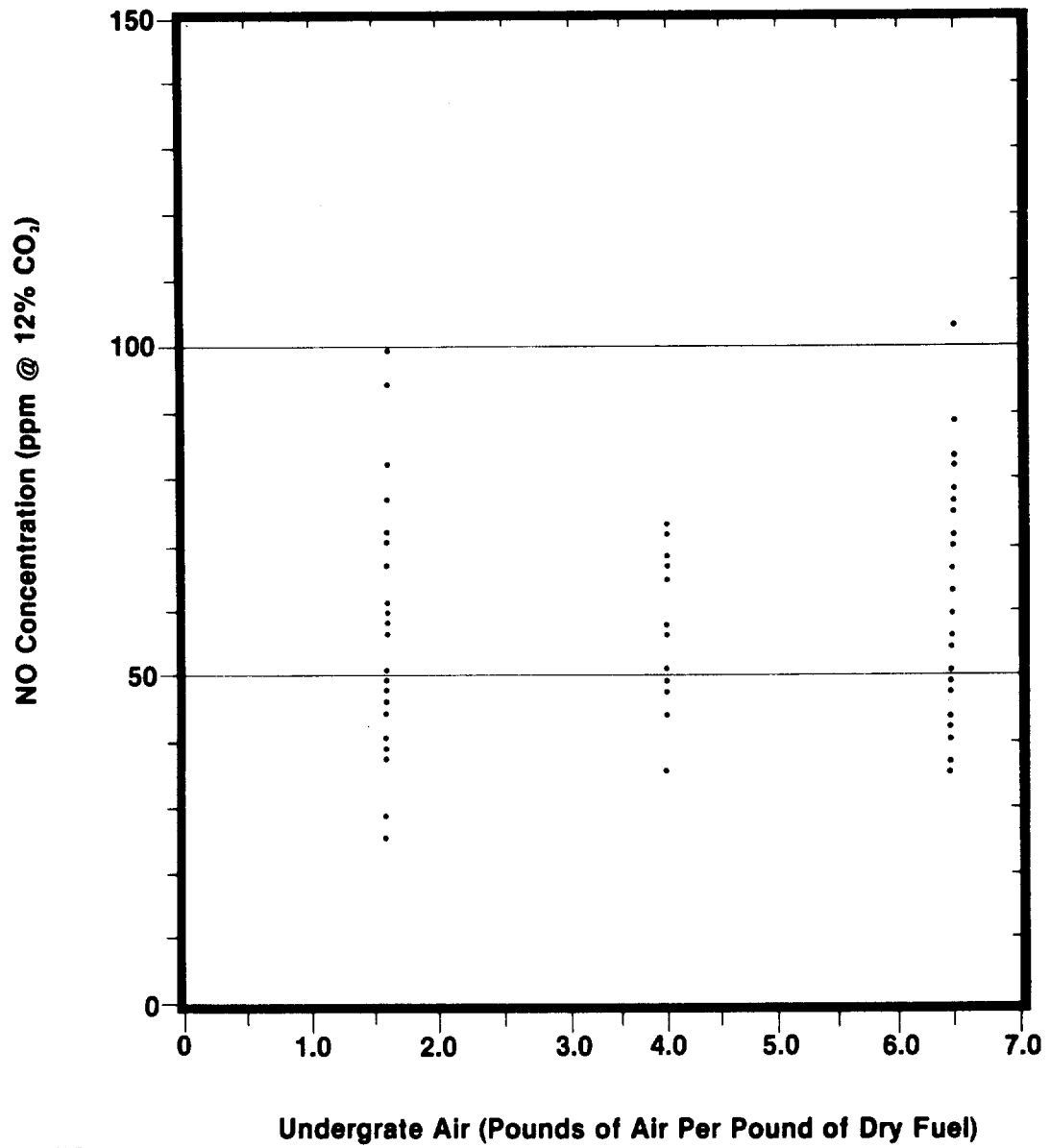
7. Effect of Undergrate Air Flow Rate (UAIR)

The effect of the undergrate air flow rate on the NO concentration is shown in figure 13. There appears to be a very slight increase in the NO concentration with increasing undergrate air flow rate. The coefficient of correlation is 0.0216.



Fuel Moisture Content Per NO Concentration

FIGURE 12



Undergrate Air Per NO Concentration

FIGURE 13

experimental projects, other errors did occur. However, they are considered to be of relatively minor significance in the overall results of this study.

#### 1. Errors in Measurement of Temperature

The temperature measurements inside the combustion chamber were subject to significant errors. The design of the thermocouple protective housing or shield was such as to allow radiation losses of large magnitude to the cold walls of the water wall lined combustion chamber. Thus, while heat energy was input to the temperature probe through convention and radiation from the combustion area, heat losses by radiation from the thermocouple to the cold walls of the combustion chamber reduced the indicated temperatures substantially. The higher the actual temperatures of the exhaust gases, the greater were the losses due to radiation away from the temperature probe. It is possible that when the probe indicated temperatures in the range of 900 to 1100 degrees F, the actual temperatures were in a range of 1500 to 2000 degrees F. This is substantiated by the fact that theoretical temperatures for wood combustion range up to 3300 degrees F for dry wood at

volume of 237 acf (350 degree F) per 1.885 lb wet bark at 47 percent water content (1 lb dry bark plus .885 lb water). The stack gas contains 20 percent  $H_2O$  by volume at this temperature.

5. The exhaust gas volume corrected to 12 percent  $CO_2$  and 70 degrees F is 168 scf per 1.885 lb wet bark or 71.3 standard dry cubic feet per pound of wet bark (dry gas basis).
6. One pound mole of gas occupies 387 scf volume at 70 degrees F and 1 atmosphere.

NO emission factor =

$$\frac{(57 \text{ ppm})(71.3 \text{ scf/lb})(30 \text{ lb NO/lb mole NO})(2,000 \text{ lb/T})}{(10^6 \text{ parts})(387 \text{ scf/lb mole gas})}$$

$$= 0.63 \text{ lb NO/ton wet bark}$$

During the design of the experiment, it was expected that virtually all of the total nitrogen oxides generated from wood combustion would be in the form of NO. According to Monitor (1976), 95 percent of the  $NO_x$  is NO by thermal equilibrium at 1,340 degrees F. However, it was not known exactly how much  $NO_2$  would

be generated in the bark fired boiler or if some  $\text{NO}_2$  would be lost in the sampling train due to the condensation of water from the combustion gases. Malte (1978) warns of this potential in his recent work, although Kramlich (1978) states that no variation in the concentration of  $\text{NO}_2$  or  $\text{NO}$  was noted as water condensed on the inside walls of his sample line in an ice bath trap sampling train similar to the one used in this study.

Cato (1974) also mentions that, since  $\text{NO}_2$  is soluble in water, it may be lost in a "wet" sample train. It was assumed that, since  $\text{NO}$  is much less soluble than  $\text{NO}_2$ , much less  $\text{NO}$  would be lost than  $\text{NO}_2$  in the train. However, although Cato postulates reactions in which up to one mole of  $\text{NO}$  per mole of  $\text{NO}_2$  may be lost to the water, his actual data do not indicate that a significant quantity of  $\text{NO}$  is, in fact, lost in this type of sample train. He reports that simultaneous measurements were made of 168 sample runs of  $\text{NO}$  from boilers with cold (wet) and hot (dry) sample lines, and the results were that cold line measurements were 2 percent lower than hot line readings. The estimated error was  $\pm 3$  ppm.

To verify the concentration of NO, NO<sub>2</sub>, and NO<sub>x</sub>, measurements were made using a Monitor Labs Model 8440E chemiluminescence analyzer modified to read stack concentrations. The instrument was modified using special instructions and flow regulators supplied by the instrument manufacturer (Williams, 1978). During a separate portion of the pilot plant experimental runs, NO<sub>2</sub> concentrations averaging 10 percent of the NO concentration were measured over a wide range of combustion variables, similar to those used in this study.

This finding is supported by the author (1976), when grab samples from hog fuel boilers held in glass flasks, aluminized mylar, the Tedlar bags at ambient temperatures for 30 to 40 minutes and analyzed with a University of Washington TECO chemiluminescence NO<sub>x</sub> analyzer showed an NO<sub>2</sub> concentration of 17 percent of the total NO<sub>x</sub> concentration. It is assumed that oxidation of NO to NO<sub>2</sub> in the containers accounted for the higher NO<sub>2</sub> concentrations. This is further supported by EPA (1977) in which they state that 90 percent of the NO<sub>x</sub> emitted from an industrial boiler is in the form of NO, the remaining 10 percent being NO<sub>2</sub>. Cato (1974) indicates that 5 percent of the NO<sub>x</sub> is NO<sub>2</sub> from some fifty boilers tested.

The EPA (1977) emission factor for  $\text{NO}_x$  emitted from wood combustion is given as 10 pounds  $\text{NO}_2$  for each ton of wood (wet basis) burned. In order to compare the measurements of  $\text{NO}_x$  made during this study, the value of 0.63 pound of NO per ton of wet bark fuel burned can be converted to 1.1 pounds of  $\text{NO}_x$  per ton of wet fuel burned. This figure incorporates the measurements that show the  $\text{NO}_2$  concentration was 10 percent of the measured NO concentration, and the reporting of the NO concentration as  $\text{NO}_2$  on a mass basis.

The conversion calculation is as follows:

$\text{NO}_x$  emission factor =

$$\left[ \frac{0.63 \text{ lb NO}}{\text{ton wet bark}} \right] \left[ \frac{1 \text{ mole NO}}{30 \text{ lb NO}} \right] \left[ \frac{1.1 \text{ moles NO}_2}{\text{mole NO}} \right] \left[ \frac{46 \text{ lb NO}_2}{\text{mole NO}_2} \right]$$

$\text{NO}_x$  Emission Factor = 1.1 lb  $\text{NO}_x$  as  $\text{NO}_2$  per ton of wet bark burned.

B. EMISSION FACTOR USAGE

This work suggests that hog fuel fired boilers are not a great a source of nitrogen oxides as previously supposed. As a result of the work done to date on NO<sub>x</sub> emission and wood combustion, the author has been contacted by EPA and asked to provide data for use in modifying the existing EPA emission factor of 10 pounds of NO<sub>x</sub> per ton of wood waste burned for hog fuel boilers. The author has received inquiries from both Region X (Seattle) and Region IV (Atlanta) regarding this work. Based on the data gathered to date, both regional offices of EPA are now using an emission factor of 1.5 pounds of NO<sub>x</sub> per ton of hog fuel burned.

C. DEVELOPMENT OF NO PREDICTIVE MODEL

In most research problems where regression analysis is applied, more than one independent variable is needed in the regression model. The complexity of most scientific mechanisms is such that, in order to be able to predict an important response, a multiple regression model is needed. When the model coefficients are linear, it is called a multiple linear regression model (Walpole, 1972). The use of multiple linear regression techniques is well suited to analyze

data as collected in this experiment. This technique determines the coefficients of a linear equation of the form:

$$y = b_0 + b_1x_1 + b_2x_2 \dots + b_nx_n,$$

where  $x_1, x_2 \dots x_n$  are the  $n$  independent variables,  $y$  is the dependent variable and  $b_0, b_1 \dots b_n$  are constant coefficients. The calculated  $y$  values approximate the measured NO concentrations in the best least squares fit and an overall multiple correlation coefficient can be calculated.

The general method of solution of the coefficients is stepwise. At each step one variable is added to the regression equation. The variable added is the one which makes the greatest reduction in the error sum of squares, and it is the variable which has the highest partial correlation with the dependent variable. As each variable is analyzed for addition to the equation, its significance is determined, and the most significant variables are processed first. With the use of this technique, it is possible to arrange the measured test parameters to give a prediction of nitrogen oxide concentrations. The significance of

the variables is in the same order as their appearance in the equation.

The best linear equation of the NO concentrations can be shown as:

$$\text{NO ppm} = 0.0623 \text{ RATE} + 0.0935 \text{ TEMP} + 0.259 \text{ XAIR} - 6.29 \text{ SIZE} + 60.5 \text{ H}_2\text{O} + 0.678 \text{ UAIR} - 145$$

Where:

RATE = rate of fuel feed in pounds per hour

XAIR = total air to fuel ratio in percent excess air

TEMP = maximum combustion temperature in degrees F

UAIR = Undergrate air in pounds of air per pound of fuel

H<sub>2</sub>O = percent moisture in the fuel (wet basis)

SIZE = size of fuel burned (1 = fine, 2 = coarse)

The equation has a multiple correlation coefficient of 0.861. This means that 74.1 percent of the variation in NO concentration can be explained by the linear regression model. The increase in correlation coefficient falls off rapidly after the first three

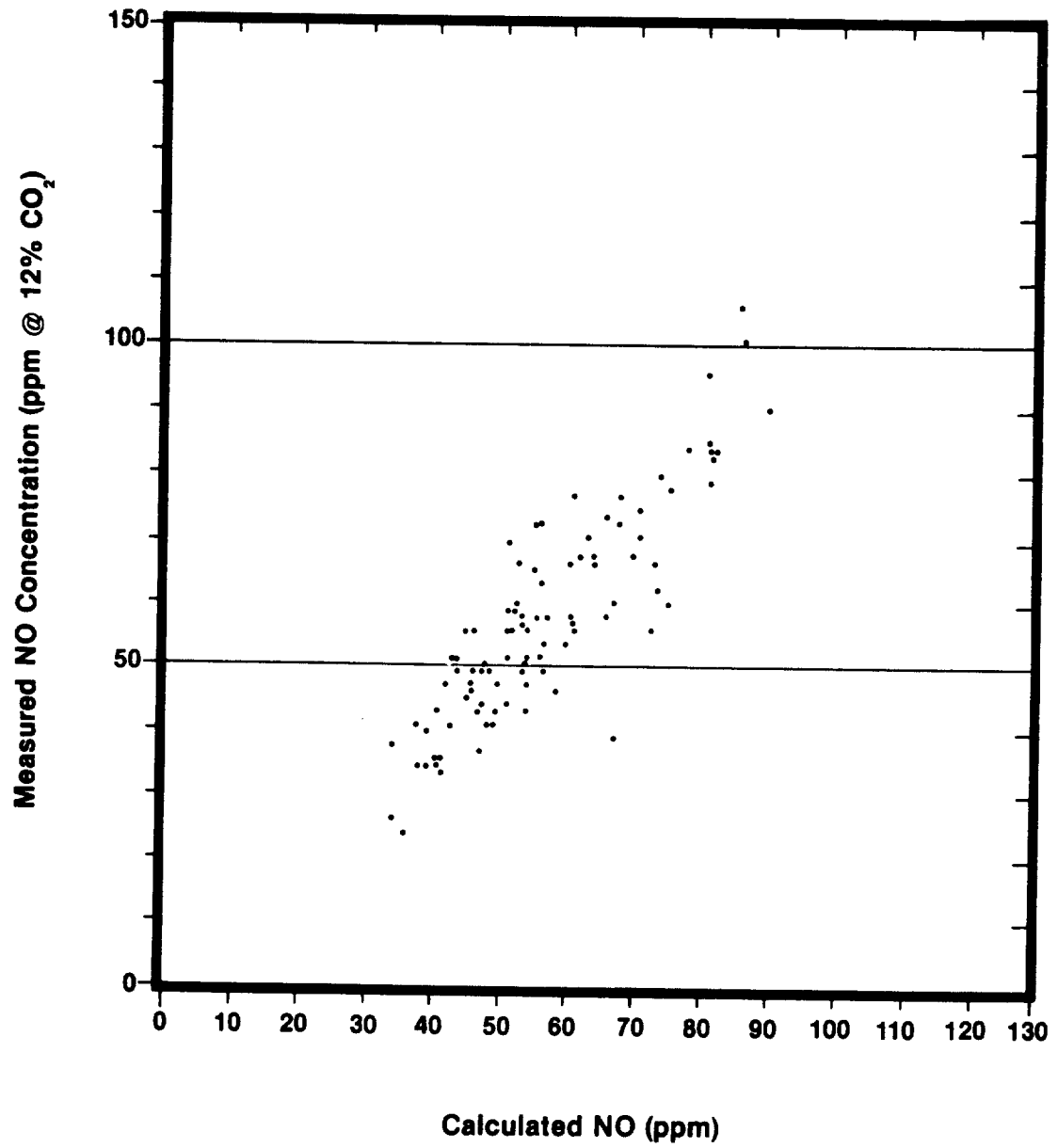
variables are evaluated. The calculated versus measured NO concentrations are shown in figure 14.

In reviewing the literature, the author was able to find only one other use of this technique in predicting NO<sub>x</sub> emissions from combustion parameters for any fuel. The reported instance was for NO<sub>x</sub> emissions from coal combustion (Cato, 1976), and the NO<sub>x</sub> emissions were calculated as a function of three variables: fuel nitrogen content, fraction of theoretical air, and heat release per unit heat/absorption area.

#### D. EXPERIMENTAL ERRORS

The errors involved in any measurement are usually of two types, random or reading errors and biased or equipment errors. Random errors depend on the sensitivity of the instruments and may be minimized by careful reading of the scales. Biased errors usually shift the experimental result in magnitude with respect to the real value and may be reduced by careful calibrations and experimental procedure.

Each potential major source of error in this experiment is discussed in this section. As with all



Calculated NO Per Measured NO Concentration

FIGURE 14

experimental projects, other errors did occur. However, they are considered to be of relatively minor significance in the overall results of this study.

#### 1. Errors in Measurement of Temperature

The temperature measurements inside the combustion chamber were subject to significant errors. The design of the thermocouple protective housing or shield was such as to allow radiation losses of large magnitude to the cold walls of the water wall lined combustion chamber. Thus, while heat energy was input to the temperature probe through convention and radiation from the combustion area, heat losses by radiation from the thermocouple to the cold walls of the combustion chamber reduced the indicated temperatures substantially. The higher the actual temperatures of the exhaust gases, the greater were the losses due to radiation away from the temperature probe. It is possible that when the probe indicated temperatures in the range of 900 to 1100 degrees F, the actual temperatures were in a range of 1500 to 2000 degrees F. This is substantiated by the fact that theoretical temperatures for wood combustion range up to 3300 degrees F for dry wood at

20 percent excess air, and are about 2100 degrees F for 50 percent moisture content wood (wet basis) fired at 50 percent excess air (Miller, 1951).

While it may seem relatively simple to construct or purchase an adequately shielded thermocouple to solve these measurement problems, in actual practice such a device is not available in the marketplace and is extremely difficult to construct (Tuttle, 1979, and Schubert, 1973).

## 2. Errors in Measurement of Gas Flow Rate

Two distinct methods were used to determine gas flow rates in the test facility. The first was to use calibrated venturi flowmeters and differential pressure gages coupled with temperature indicators to determine the flow rates of air input to the system. The venturis were calibrated using a standard Pitot tube and crossed diameter traverses. Errors in flow measurement using these procedures are likely to be less than 5 percent.

The second method of determining gas flow rates was to determine the levels of excess air entering

the combustion chamber based on flue gas analyses for oxygen determination. Knowing the level of oxygen in the flue gas allows a direct calculation of the excess air based on the amount of dry fuel being burned at any time. Calculation of total gases entering the combustion chamber is then possible provided that an accurate measure of the incoming dry fuel feed rate is available.

Errors arise in carrying out these calculations. The rate of fuel feed, while it was reasonably constant over a time duration of 5 minutes, did experience fluctuations of small time duration (30 - 45 seconds). These fluctuations in fuel feed rate resulted in fluctuation in the amount of oxygen present in the flue gases. The use of a continuous oxygen analyser tended to damp out these fluctuations and provide data for a calculation based on average levels much longer than the fuel feed fluctuations. It is expected that the average levels of excess air were determined with a level of accuracy estimated to be within 5 percent (Junge, 1977).

### 3. Errors in Measurement of Fuel Flow Rates

The rate of fuel input to the system was measured in terms of dry pounds of fuel input per hour. To achieve this, the fuel density, moisture content, and feed rate in terms of cubic feet per hour on a wet basis were determined.

Fuel density was determined by measuring the time required to collect one full office type waste paper can. The volume of the can was easily established by geometry. The time required to fill the can from the screw feed conveyor was measured at least three times. It is felt that accuracy to within 1 percent was achieved in this process.

Fuel moisture content was determined by small grab samples analyzed in the laboratory by the use of a drying oven and standard laboratory scales. Weighing and drying procedures are well standardized and relatively accurate (within 1 percent). The errors which may occur will stem principally from the difficulties of obtaining representative samples of the fuel. The samples were all chosen at random from the fuel pile,

but a question remains as to their degree of being truly representative. It is estimated that fuel moisture determinations are likely to be within 2 percent of actual values.

Fuel feed rate was determined by calibrating the feed rate of the screw conveyor prior to the actual experiment and then carrying out calibration checks during the experiment. Appropriate replications of the calibration method were used to ensure statistical accuracy; however, the time framework for the tests was relatively short (1-2 minutes) per check. In carrying out the experimental test runs, the time length of each test varied from 10 to 40 minutes. It was assumed that the flow rate through the screw conveyor remained constant during this time. It is likely that errors in fuel feed rate of up to 2 percent are possible.

In combining the sources of all errors in measurement of the flow rate of dry fuel to the combustion test facility, it appears that significant errors were likely to occur and that these combined errors may be as much as 5 percent from the true values of fuel flow rates in dry pounds per hour (Junge, 1977).

#### 4. Errors in Measurement of NO.

Many compounds, both gases and liquids, absorb light. Generally a compound will absorb light of a particular wavelength and not light of some other wavelength. For instance, nitrogen dioxide gas absorbs visible light in the blue region (4000 to 5000 angstroms) but not in the yellow and red regions (5000 to 7000 Å). For this reason the gas appears yellow-brown.

The absorbing power of a particular gas is defined through a physical constant called the absorption coefficient,  $a(\lambda)$  which is a function of wavelengths where the gas does not absorb and becomes greater than zero at wavelengths that absorption occurs.

Light of intensity  $I_0$  of a particular wavelength  $\lambda$  can be directed through a cell of length  $b$ . Gas, inside the cell, at concentration  $c$  absorbs some of the light, and that not absorbed exits at intensity  $I$ . This exiting intensity is

predictable using the following formula, known as Beer's Law:

$$I(\lambda) = I_0(\lambda) e^{-a(\lambda)bc}$$

Where:

$I_0$  = Incident Intensity

$I$  = Exiting Intensity

$c$  = Concentration of Absorbing Gas

$b$  = Pathlength through the Gas

$a(\lambda)$  = Absorption Coefficient of the Gas

If the absorption coefficient  $a(\lambda)$  is known, the concentration  $c$  of the gas in the cell can be determined just by measuring the incident and exiting intensities. That is:

$$c = \frac{1}{a(\lambda)b} \ln \frac{I_0}{I}$$

This characteristic of absorbing radiation is a function of wavelength, as a particular gas will absorb only radiation of particular wavelengths.

There are two deficiencies in using a radiation absorption sensing system as a gas analyzer. Sensitivity is often limited, and such a gas analyzer is open to interference by a gas other than that being measured. If another gas is present in the sample cell which absorbs light at the wavelength the measured gas does, it is impossible to determine which ~~gas~~ <sup>gas</sup> is being detected and measured.

Many compounds absorb just over narrow intervals of wavelengths. These absorption bands can be relatively narrow, about 10 to 30 angstroms wide. The locations of these absorption bands are intimately tied to the geometry of the molecule. Therefore, no two molecules exhibit the same pattern of absorption bands. The fact that a compound exhibits a narrow absorption band at a particular wavelength is, therefore, a unique physical property of that compound. This property is more than the fact that it absorbs radiation at that wavelength for it requires that it does not absorb as much as 10 angstroms to either side of this wavelength. Second Derivative Spectroscopy is a method for detecting this property of narrow band absorption.

The Lear Siegler SM 1000 analyzer is a second derivative analyzer, that is, it measures the  $d^2(I/I_0)/d\lambda^2$  of the absorption of NO. Derivative spectroscopy is concerned with the shape characteristics, rather than with the basic intensity changes, of a spectral absorption distribution. The slope and curvature characteristics are often quite large, very specific, and largely independent of intensity. Because these shape characteristics are large, yet specific to individual compounds, extremely complex separations of component gases are possible, while the sensitivity over conventional spectrometric techniques is increased by several decades. The manufacturer's specifications indicate that it should be accurate to within 5 percent of full scale.

A potential problem exists in the use of the sampling train that condensed water from the stack gases, thereby providing for a possible reaction of NO and NO<sub>2</sub> with the water which would lower the recorded NO<sub>x</sub> values. However, Cato (1974) and Kramlich (1978) found no variation in NO or NO<sub>2</sub> in work with a similar sampling train (see section IV B). Additionally, the train was designed to provide a much higher flowrate than

was needed by the NO analyzer through the use of a bypass, and the water condensed was continually removed by a peristaltic pump. However, it is expected that the NO measurements may be biased slightly on the low side because of possible sample loss through combination with water in the sample train, approximately 2 percent.

## VII. CONCLUSIONS

- A.  $\text{NO}_x$  emissions from Douglas fir bark combustion are primarily related to rate of fuel feed, combustion temperature, and total air to fuel ratio. To a much lesser degree, there is a slight increase in  $\text{NO}_x$  emissions with increasing undergrate air flow rate, and slight decrease in  $\text{NO}_x$  emissions with increasing fuel size. There appears to be little correlation of  $\text{NO}_x$  emissions with fuel moisture content.
- B.  $\text{NO}_x$  emissions from the combustor are primarily NO.  $\text{NO}_2$  values are about 10 percent of the measured NO concentrations.
- C. An emission factor of 1.1 pounds of  $\text{NO}_x$  per ton of wet hog fuel burned was developed. The current EPA emission factor for wood waste fired boilers is about nine times as high as the one developed by this study.
- D. A predictive model was developed which calculates values of NO as a function of boiler operating parameters. This model may be used to estimate NO emissions from other similar boilers under varying operating conditions.

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## **APPENDIX A**

### **EXPERIMENTAL DATA**

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TABLE 4

Test No.	Fuel Size	Fuel Rate	Fuel % H <sub>2</sub> O	Under Grate Air	Percent Excess Air	Max. Temp.	NO ppm @12% CO <sub>2</sub>	NO ppm RAW
1	coarse	450	.494	6.5	80	1,320	41	38
2	coarse	500	.494	6.5	80	1,320	58	53
3	coarse	550	.494	6.5	80	1,380	55	50
4	coarse	500	.494	6.5	80	1,400	53	49
5	coarse	450	.494	6.5	80	1,310	41	38
6	coarse	475	.494	1.5	30	1,320	27	35
7	coarse	500	.494	1.5	30	1,320	25	32
8	coarse	525	.494	1.5	30	1,340	40	51
9	coarse	500	.494	1.5	30	1,300	38	48
10	coarse	495	.494	6.5	30	1,340	36	46
11	coarse	500	.494	6.5	30	1,340	36	46
12	coarse	505	.494	6.5	30	1,340	34	43
13	coarse	500	.494	6.5	30	1,320	35	44
14	coarse	575	.494	4.0	55	1,300	43	43
15	coarse	600	.494	4.0	55	1,300	49	52
16	coarse	625	.494	4.0	55	1,360	57	60
17	coarse	600	.494	4.0	55	1,360	43	45
18	coarse	595	.494	4.0	55	1,315	43	45
19	coarse	600	.494	4.0	55	1,330	44	46
20	coarse	605	.494	4.0	55	1,350	49	52
21	coarse	600	.494	4.0	55	1,360	47	50
22	coarse	650	.494	6.5	30	1,400	71	91
23	coarse	700	.494	6.5	30	1,420	75	95
24	coarse	750	.494	6.5	30	1,460	75	95
25	coarse	700	.494	6.5	30	1,420	75	95
26	coarse	650	.494	1.5	80	1,480	65	60
27	coarse	700	.494	1.5	80	1,500	82	75
28	coarse	750	.494	1.5	80	1,555	98	90
29	coarse	700	.494	1.5	80	1,530	93	85
30	coarse	605	.494	4.0	55	1,355	50	53
31	coarse	600	.494	4.0	55	1,330	51	54
32	coarse	595	.494	4.0	55	1,340	55	58
33	coarse	575	.494	4.0	55	1,410	57	60
34	coarse	600	.494	4.0	55	1,430	57	60
35	coarse	625	.494	4.0	55	1,345	55	58
36	fine	650	.539	1.5	30	1,385	65	83
37	fine	700	.539	1.5	30	1,430	71	90
38	fine	750	.539	1.5	30	1,480	76	97
39	fine	700	.539	1.5	30	1,380	69	88
40	fine	675	.539	6.5	30	1,250	59	75
41	fine	700	.539	6.5	30	1,220	58	74
42	fine	725	.539	6.5	30	1,260	53	67
43	fine	700	.539	6.5	30	1,270	51	65
44	fine	695	.539	6.5	80	1,160	46	42
45	fine	700	.539	6.5	80	1,040	49	45

Test No.	Fuel Size	Fuel Rate	Fuel % H <sub>2</sub> O	Under Grate Air	Percent Excess Air	Max. Temp.	NO ppm @12% CO <sub>2</sub>	NO ppm RAW
46	fine	705	.539	6.5	80	1,020	49	45
47	fine	700	.539	6.5	80	1,110	51	47
48	fine	550	.539	6.5	30	1,340	56	72
49	fine	500	.539	6.5	30	1,270	51	65
50	fine	450	.539	6.5	30	1,240	41	57
51	fine	500	.539	6.5	30	1,240	43	55
52	fine	550	.539	1.5	80	1,260	71	65
53	fine	500	.539	1.5	80	1,250	55	50
54	fine	450	.539	1.5	80	1,240	37	34
56	coarse	550	.369	4.0	55	1,500	49	52
57	coarse	500	.369	4.0	55	1,380	47	50
58	coarse	450	.369	4.0	55	1,400	35	37
59	coarse	450	.369	6.5	30	1,420	35	48
60	coarse	500	.369	6.5	30	1,440	41	52
61	coarse	550	.369	6.5	30	1,480	47	60
62	coarse	500	.369	6.5	30	1,470	47	60
63	coarse	495	.369	1.5	80	1,350	49	48
64	coarse	500	.369	1.5	80	1,360	55	50
65	coarse	505	.369	1.5	80	1,370	55	50
66	coarse	500	.369	1.5	80	1,340	51	47
71	coarse	695	.369	6.5	80	1,580	83	76
72	coarse	700	.369	6.5	80	1,580	82	75
73	coarse	705	.369	6.5	80	1,580	82	75
74	coarse	700	.369	6.5	80	1,580	81	74
75	fine	695	.463	6.5	30	1,440	39	50
76	fine	700	.463	6.5	30	1,510	78	100
79	fine	695	.463	1.5	80	1,190	57	52
80	fine	700	.463	1.5	80	1,420	59	54
81	fine	705	.463	1.5	80	1,400	61	56
82	fine	700	.463	1.5	80	1,420	59	54
83	fine	650	.463	1.5	30	1,440	56	72
84	fine	700	.463	1.5	30	1,460	57	73
85	fine	750	.463	1.5	30	1,440	59	75
86	fine	700	.463	1.5	30	1,440	66	84
87	fine	650	.463	6.5	80	1,390	55	50
88	fine	700	.463	6.5	80	1,450	77	70
89	fine	750	.463	6.5	80	1,510	88	80
90	fine	700	.463	6.5	80	1,500	103	94
91	fine	650	.463	4.0	55	1,450	66	70
92	fine	600	.463	4.0	55	1,440	72	76
93	fine	550	.463	4.0	55	1,320	68	72
95	fine	505	.463	1.5	30	1,370	45	58
96	fine	500	.463	1.5	30	1,380	46	59
97	fine	495	.463	1.5	30	1,400	44	57
98	fine	500	.463	1.5	30	1,400	50	64
99	fine	550	.463	6.5	80	1,440	69	63

<u>Test No.</u>	<u>Fuel Size</u>	<u>Fuel Rate</u>	<u>Fuel % H<sub>2</sub>O</u>	<u>Under Grate Air</u>	<u>Percent Excess Air</u>	<u>Max. Temp.</u>	<u>NO ppm @12% CO<sub>2</sub></u>	<u>NO ppm RAW</u>
100	fine	500	.463	6.5	80	1,400	65	60
101	fine	450	.463	6.5	80	1,350	62	57
102	fine	500	.463	6.5	80	1,280	65	60
103	fine	550	.463	4.0	55	1,360	64	68
104	fine	600	.463	4.0	55	1,400	66	70
105	fine	650	.463	4.0	55	1,460	73	77

## APPENDIX B

### MODEL PREDICTION OF DATA SETS

TABLE 5

<u>No.</u>	<u>Actual NO Value</u>	<u>Predicted NO Value</u>	<u>No.</u>	<u>Actual NO Value</u>	<u>Predicted NO Value</u>
1	25	36	50	55	46
2	27	34	51	55	12
3	34	41	52	56	53
4	35	39	53	56	61
5	35	41	54	57	55
6	35	38	55	57	57
7	36	41	56	57	60
8	36	41	57	57	53
9	37	47	58	57	66
10	38	34	59	58	52
11	39	67	60	58	51
12	40	39	61	59	52
13	41	49	62	59	75
14	41	48	63	59	75
15	41	37	64	59	67
16	41	43	65	61	73
17	43	47	66	62	56
18	43	54	67	64	55
19	43	49	68	65	73
20	43	41	69	65	60
21	44	51	70	65	64
22	44	47	71	65	53
23	45	45	72	66	64
24	46	58	73	66	70
25	46	46	74	66	62
26	47	54	75	68	51
27	47	42	76	69	63
28	47	50	77	69	71
29	47	46	78	71	56
30	49	48	79	71	67
31	49	53	80	71	55
32	49	47	81	72	66
33	49	46	82	73	71
34	49	56	83	75	61
35	49	44	84	75	68
36	50	54	85	75	61
37	50	48	86	76	75
38	51	51	87	77	81
39	51	56	88	74	74
40	51	54	89	81	81
41	51	43	90	82	78
42	51	43	91	82	81
43	53	60	92	82	82
44	53	56	93	83	81
45	55	61	94	88	90
46	55	52	95	93	81
47	55	54	96	98	86
48	55	51	97	103	86
49	55	45			

## APPENDIX C

### COMPUTER DATA ANALYSIS

## C. COMPUTER DATA ANALYSIS

## \*\*\*\*\* MULTIPLE LINEAR REGRESSION \*\*\*\*\*

7 VARIABLES; VARIABLE: NO IS DEPENDENT

STANDARD ERROR OF Y = 15.56372

## \*\*\*\*\* CORRELATION MATRIX \*\*\*\*\*

VAR.	SIZE	RATE	H2O	VAIR	XAIR	TEMP	NO
SIZE	1.0000						
RATE	-0.2576	1.0000					
H2O	-0.3572	0.1871	1.0000				
VAIR	0.0671	-0.0046	0.0047	1.0000			
XAIR	0.0480	0.0695	-0.1873	0.0034	1.0000		
TEMP	0.2386	0.2428	-0.5471	-0.0951	-0.0074	1.0000	
NO	-0.1988	0.6448	-0.0656	0.0216	0.3398	0.5488	1.0000

STEP NO. 1  
 ENTERING VARIABLE: RATE  
 F-LEVEL 67.61951 WITH PROB. 0.0000  
 STANDARD ERROR OF ESTIMATE 11.95811  
 COEFFICIENT OF DETERMINATION = 0.4158143  
 COEFFICIENT OF MULTIPLE REGRESSION = 0.6448366  
 INCREASE IN COEFFICIENT OF DETERMINATION = 0.4158143  
 CONSTANT -6.9361

VARIABLE	COEFFICIENT	STD ERROR OF COEFF.
RATE	0.1065519	0.1295762E-01

STEP NO. 2  
 ENTERING VARIABLE: TEMP  
 F-LEVEL 36.52872 WITH PROB. 0.0000  
 STANDARD ERROR OF ESTIMATE 10.20167  
 COEFFICIENT OF DETERMINATION = 0.5792998  
 COEFFICIENT OF MULTIPLE REGRESSION = 0.7611175  
 INCREASE IN COEFFICIENT OF DETERMINATION = 0.1634855  
 CONSTANT -82.028

VARIABLE	COEFFICIENT	STD ERROR OF COEFF.
RATE	0.8982870E-01	0.1139540E-01
TEMP	0.6200150E-01	0.1025853E-01

```

STEP NO. 3
ENTERING VARIABLE: XAIR
F-LEVEL 26.60958 WITH PROB. 0.0000
STANDARD ERROR OF ESTIMATE 9.043831
COEFFICIENT OF DETERMINATION = 0.6728931
COEFFICIENT OF MULTIPLE REGRESSION = 0.8203006
INCREASE IN COEFFICIENT OF DETERMINATION = 0.9359329E-01
CONSTANT -93.198

VARIABLE COEFFICIENT STD ERROR OF COEFF.
RATE 0.8598721E-01 0.1012948E-01
XAIR 0.2175831 0.4217996E-01
TEMP 0.6317738E-01 0.9097087E-02

```

STEP NO. 4  
 ENTERING VARIABLE: SIZE  
 F-LEVEL 12.56406 WITH PROB. 0.0006  
 STANDARD ERROR OF ESTIMATE 8.529090  
 COEFFICIENT OF DETERMINATION = 0.7121971  
 COEFFICIENT OF MULTIPLE REGRESSION = 0.8439177  
 INCREASE IN COEFFICIENT OF DETERMINATION = 0.3930404E-01  
 CONSTANT -90.572

VARIABLE	COEFFICIENT	STD ERROR OF COEFF.
SIZE	-6.734386	1.899910
RATE	0.7374285E-01	0.1015833E-01
XAIR	0.2290000	0.3990943E-01
TEMP	0.7358508E-01	0.9067861E-02

STEP NO. 5  
 ENTERING VARIABLE: H2O  
 F-LEVEL 6.959416 WITH PROB. 0.0098  
 STANDARD ERROR OF ESTIMATE 8.265583  
 COEFFICIENT OF DETERMINATION = 0.7326437  
 COEFFICIENT OF MULTIPLE REGRESSION = 0.8559461  
 INCREASE IN COEFFICIENT OF DETERMINATION = 0.2044663E-01  
 CONSTANT -138.67

VARIABLE	COEFFICIENT	STD ERROR OF COEFF.
SIZE	-5.978906	1.863350
RATE	0.6357464E-01	0.1057215E-01
H2O	58.27590	22.09035
XAIR	0.2578455	0.4019235E-01
TEMP	0.9093786E-01	0.1097687E-01

STEP NO. 6  
 ENTERING VARIABLE: VAIR  
 F-LEVEL 3.039870 WITH PROB. 0.0846  
 STANDARD ERROR OF ESTIMATE 8.174471  
 COEFFICIENT OF DETERMINATION = 0.7413790  
 COEFFICIENT OF MULTIPLE REGRESSION = 0.8610337  
 INCREASE IN COEFFICIENT OF DETERMINATION = 0.8735269E-02  
 CONSTANT -145.01

VARIABLE	COEFF.	STD.ERR. OF COEFF.	STANDARDIZED COEFFICIENT	T-VALUE
SIZE	-6.289	1.851	-0.2029796	-3.397
RATE	0.6227E-01	0.1048E-01	0.3768601	5.941
H2O	60.46	21.88	0.2033485	2.763
VAIR	0.6775	0.3886	0.9459050E-01	1.744
XAIR	0.2594	0.3976E-01	0.3657484	6.525
TEMP	0.9352E-01	0.1096E-01	0.6286715	8.536

APPENDIX D

PUBLICATION OF RESEARCH

# Nitrogen oxide emissions from a pilot plant spreader-stoker bark-fired boiler

## ABSTRACT

*The nitrogen oxide concentration in the gases exhausted from a pilot plant spreader-stoker bark-fired boiler was measured as a function of fuel feed rate, excess air, combustion temperature, fuel size, underfire air, and fuel moisture content. The nitrogen oxide concentration was measured with an ultraviolet analyzer. The average concentration, corrected to 12% CO<sub>2</sub>, was 57 ppm. An NO<sub>x</sub> emission factor of 1.1 lb of NO<sub>x</sub> per ton of wet bark burned was calculated for the Douglas fir bark fuel.*

**Richard A. Kester and Michael J. Pilat**

Dept. of Civil Engineering, University of Washington, Seattle, Wash. 98115

TAPPI EDITORIAL DEPARTMENT

1 DUNWOODY PARK, ATLANTA, GA., 30341

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Wood has been used as an industrial fuel for many decades in the United States. In the early part of this century, wood was a major energy source but was gradually replaced by coal and later by petroleum. However, wood is now being reevaluated as an energy source in view of the price and availability of fossil fuels. In order to gain regulatory approval to install new wood-fired boilers, it is necessary to show by computer modeling analysis that the air contaminant emissions will not violate air quality standards. Reliable emission factors for emissions from wood combustion are necessary as inputs to the air quality models.

The objectives of this study were to investigate nitrogen emitted from a pilot plant spreader-stoker bark-fired boiler and to develop a nitrogen oxide and NO<sub>x</sub> (nitrogen oxide plus nitrogen dioxide) emission factor for Douglas fir bark combustion. The approach used in this study was to measure the nitrogen concentration in the gases exhausted from a pilot plant spreader-stoker boiler which burns up to 750 lb (340 kg) of bark per hr.

Proposed mechanisms and reaction rates for NO<sub>x</sub> formation in fossil fuel combustion have been reported by a number of authors, including Fenimore (1) and Malte (2). Wood combustion reaction rates and combustion products other than NO<sub>x</sub> have also been reported by Kreisinger (3), Prakash (4), Kester (5), and Junge (6). Galeano (7) reported an NO<sub>x</sub> emission concentration of 123 ppm for a hog fuel boiler burning a small amount of natural gas at a kraft pulp mill. Schmall (8) reported a 50–54 ppm NO<sub>x</sub> concentration in the emissions from a boiler burning hog fuel and a chipped tires and hog fuel mix. Kester (9) tested three hog fuel boilers using grab sampling techniques and reported NO<sub>x</sub> concentrations ranging from 40 to 75 ppm.

## Results

The NO concentrations were recorded at various fuel feed rates, percent excess air, underfire air flow rates, fuel size, and moisture content. The NO concentrations, corrected to 12% CO<sub>2</sub>, range from 25 to 103 ppm with the aver-

age at 57 ppm and the geometric mean at 56 ppm, as shown in Fig. 1.

### NO emission factor

An NO emission factor (lb of NO emissions per ton of wood bark burned) was developed using the NO concentrations in the exhaust gases, the exhaust gas flow rate, the rate of bark burned, and stoichiometric combustion calculations. The calculations include:

1. The average NO concentration (from Fig. 1) was 57 ppm, corrected to 12% CO<sub>2</sub> and on a dry gas basis.
2. The weighted average water content of the bark fuel for all the tests was 47% (wet basis).
3. At 50% excess air, bark combustion provides exhaust gases with 13% CO<sub>2</sub> (dry gas basis).
4. Stoichiometric combustion calculations at 50% excess air, 13% CO<sub>2</sub>, and with 47% water content bark provide an exhaust gas volume of 237 actual ft<sup>3</sup> (6.71 actual m<sup>3</sup>) at 350°F (177°C) per 1.885 lb (0.856 kg) of wet bark at 47% water content (wet basis). The stack gas contains 20% H<sub>2</sub>O by volume.
5. The exhaust gas volume corrected to 12% CO<sub>2</sub> and 70°F (21°C) is 168 std. ft<sup>3</sup> (4.75 std. m<sup>3</sup>) per 1.885 lb (0.856 kg) wet bark or 71.3 std. dry ft<sup>3</sup> (2.02 std. dry m<sup>3</sup>) per lb (0.454 kg) of wet bark (dry gas basis).
6. One pound (0.454 kg) mole of gas occupies 387 std. ft<sup>3</sup> (11.0 std. m<sup>3</sup>) volume at standard conditions.

The NO emission factor equals 57 ppm NO times 71.3 std. ft<sup>3</sup> of wet bark times 30 lb of NO/lb mole NO times 2000 lb/ton, divided by 387 std. ft<sup>3</sup>/lb mole gas times and divided by 10<sup>6</sup> parts, or, 0.63 lb of NO/ton of wet bark (0.32 of NO/kg of wet bark).

During the design of the experiment, it was expected that virtually all of the total nitrogen oxides generated from wood combustion would be in the form of NO. However, it was not known exactly how much NO<sub>2</sub> would be generated in the bark-fired boiler, or if some NO<sub>2</sub> would be lost in the sampling train due to the condensation of water from the combustion gases. Malte (10) warns of this potential in his recent work,

although Kramlich (11) states that no variation in the concentration of NO<sub>2</sub> or NO was noted as water condensed on the inside walls of his sample line in an ice bath trap sampling train similar to the one used in this study.

To verify the concentrations of NO, NO<sub>2</sub>, and NO<sub>x</sub>, measurements were made using a Monitor Labs Model 8440E chemiluminescence analyzer, modified to read stack concentrations, and calibrated with 100 and 200 ppm span gas. During a separate portion of the pilot plant spreader stoker experimental runs, NO<sub>2</sub> concentrations averaging 10% of the NO concentration were measured over a wide range of combustion variables. This finding is supported by the author's earlier work, when grab samples held in glass flasks, aluminized mylar, and Tedlar bags at ambient temperatures for 30–40 min and analyzed with a TECO chemiluminescence NO<sub>x</sub> analyzer showed an NO<sub>2</sub> concentration of 17% of the total NO<sub>x</sub> concentration. It is assumed that oxidation of NO to NO<sub>2</sub> in the bag accounted for the higher NO<sub>2</sub> concentrations.

The EPA (12) emission factor for NO<sub>x</sub> emitted from wood combustion is given as 10 lb of NO<sub>x</sub> for each ton of wood (wet) burned (5 g of NO<sub>2</sub> per kg of wet wood). In order to compare the measurements of NO<sub>x</sub> made during this study, the value of 0.63 lb of NO per ton of wet bark fuel burned can be converted to 1.1 lb of NO<sub>x</sub> per ton of wet fuel burned (0.55 g of NO<sub>x</sub> per kg of wet wood). This figure incorporates the measurements that show that the NO<sub>2</sub> concentration was 10% of the measured NO concentration and the reporting of the NO concentration as NO<sub>2</sub> on a mass basis.

The conversion calculation is as follows: NO<sub>x</sub> emission factor = (0.63 lb NO/ton of wet bark) × (1 mole of NO/30 lb of NO) × (1.1 moles of NO<sub>2</sub>/mole of NO) × (46 lb of NO<sub>2</sub>/mole NO<sub>2</sub>) = 1.1 lb NO<sub>x</sub> as NO<sub>2</sub> per ton of wet bark burned (0.55 g of NO<sub>x</sub> per kg wet bark burned)

### Summary

This study of the emission of NO from the experimental spreader-stoker boiler burning wet Douglas fir bark led to several conclusions:

1. Values of 25 to 103 ppm, with an average of 57 ppm NO, on a dry gas basis corrected to 12% CO<sub>2</sub> were recorded, shown in Fig. 1.

2. The NO<sub>x</sub> was emitted primarily as NO, with about 10% being NO<sub>2</sub>.

3. NO emissions increased with increasing fuel feed rates, as shown in Fig. 2.

4. NO emissions increased with increasing combustion temperatures, as shown in Fig. 3.

5. NO emissions increased with increasing excess air, as shown in Fig. 4.

6. NO emissions decreased slightly with increasing fuel size, as shown in Fig. 5.

7. NO emissions showed little effect of changes in moisture content or rate of underfire air, as shown in Figs. 6 and 7.

8. An emission factor of 1.1 lb of NO<sub>x</sub> per ton (0.55 g per kg) of wet fuel burned was developed.

## Experimental

### Test facilities

In this experiment, the NO concentrations were measured in the flue gases exhausting from a pilot plant spreader-stoker hog fuel boiler operating at 450–750 lb (240–340 kg) of bark per hr (bone dry weight). This wood waste combustion facility is schematically shown in Fig. 8.

The pilot plant boiler has a grate area of 10 ft<sup>2</sup> (0.92 m<sup>2</sup>). The combustion zone extends some 11 ft (3.4 m) above the grate. The pilot plant is designed to operate at up to 10 million Btu/hr (2.9 × 10<sup>6</sup> W) combustion rates. Typical tests were carried out in the 4.2 to 7.0 million Btu/hr (1.2 × 10<sup>6</sup> to 2.1 × 10<sup>6</sup> W) range. This resulted in heat release rates of 25,000 to 41,000 Btu/ft<sup>3</sup> (9.3 × 10<sup>5</sup> to 1.5 × 10<sup>6</sup> J/m<sup>3</sup>)/hr. The pilot plant was operated in the steady state in essentially the same manner as full-scale industrial hog fuel boilers.

The fuel used during this study was Douglas fir bark with analysis as shown in Table I.

### Measurement methods

**Nitric oxide.** The nitrogen oxide was measured with a Lear Seigler model SM 200 ultraviolet analyzer. The gas sample was withdrawn at a flow rate of 1 ft<sup>3</sup>/min (1.7 m<sup>3</sup>/hr) upstream of the air

preheater through the gas sample port, as shown in Fig. 8.

The NO sampling train consisted of a stainless steel probe followed by an ice bath, a coarse filter, a water trap equipped with a peristaltic water pump to drain the trap, a diaphragm gas pump, a sample line heater to raise the gas temperature 80° to 100°F (27° to 37°C), a fine filter, and on to the NO analyzer. There was a bypass for the gas sample flow rate in excess of the 400 cm<sup>3</sup>/min needed by the NO analyzer.

**Fuel feed rate.** The fuel feed rate was measured by weighing the bark fuel delivered by the screw conveyors for a measured period of time. The fuel feed rate was measured at various speeds of each screw conveyor in order to calibrate the variac and ammeters on the variable-speed electric motor. Fuel samples were dried and moisture contents measured so that fuel feed rates on a bone dry basis were known.

**Excess air.** The total flow (underfire plus overfire) to the combustion chamber was controlled using damper valves with venturi flowmeters and Magnahelix pressure differential gauges. The desired amount of combustion air flow was calculated for each given percent excess air by stoichiometric combustion calculations. The combustion air temperature was held constant at about 280°F (138°C).

The combustion air flow rate was set to run at 30, 50, and 80% excess air for the various fuel rates.

**Gas temperature.** The combustion gas temperature was measured with a shielded thermocouple (single shield) located 63 in. (163 cm) above the grate.

**Fuel size.** The fuel was prescreened into two size fractions. The coarse fuel ranged from 0.3 to 1.3 in. (8 to 32 mm) diameter, whereas the fine fuel was 0.3 in. (8 mm) and under.

**Fuel moisture content.** The fuel moisture content was measured by weighing a fuel sample before and after drying to constant weight at 219°F (104°C).

**Underfire air flow rate.** The underfire air flow rate was measured using a venturi flowmeter and differential pressure gauge. The desired underfire air flow rate was reported in lb of air per lb of bone dry fuel and ranged from 1.5 to 6.5. With the percentages of carbon, hydrogen, and oxygen given in Table I for the Douglas fir bark fuel, stoichiometric combustion calculations

show that 6.7 lb (3 kg) of air are required for complete combustion of 1 lb (0.454 kg) of fuel.

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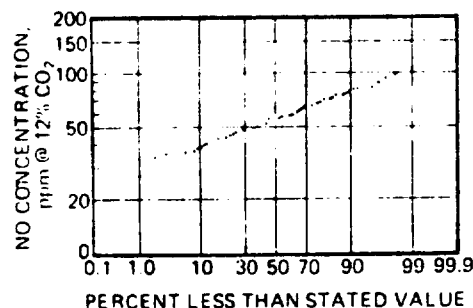
### KEYWORDS

Nitrogen oxides  
Bark  
Hogged fuel  
Combustion  
Boilers  
Emission

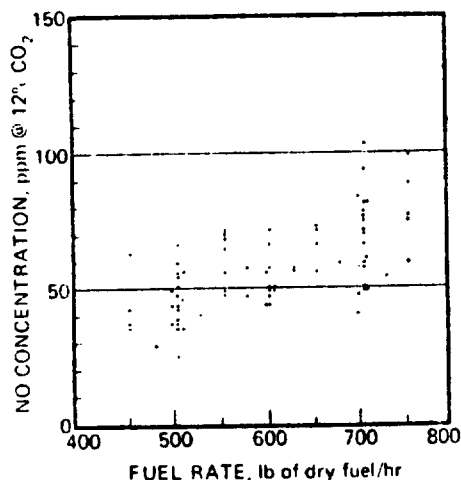
### I. Analysis of Douglas fir bark

Weight (dry basis), %	
Proximate analysis	
Volatile matter	71.0
Fixed carbon	28.0
Ash	1.0
Ultimate analysis	
Hydrogen	6.1
Carbon	54.1
Nitrogen	0.2
Oxygen	38.8
Sulfur	0.0
Ash	1.0

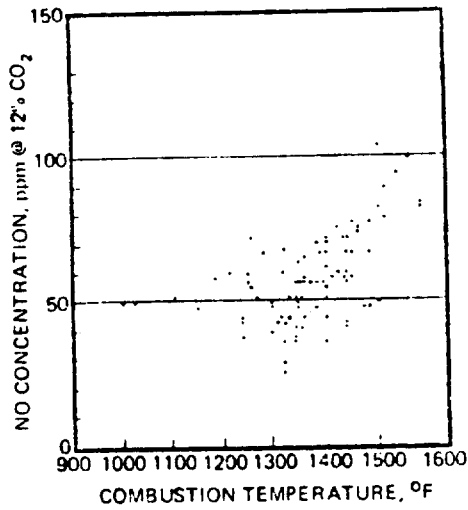
### 1. Distribution of measured nitric oxide concentrations.



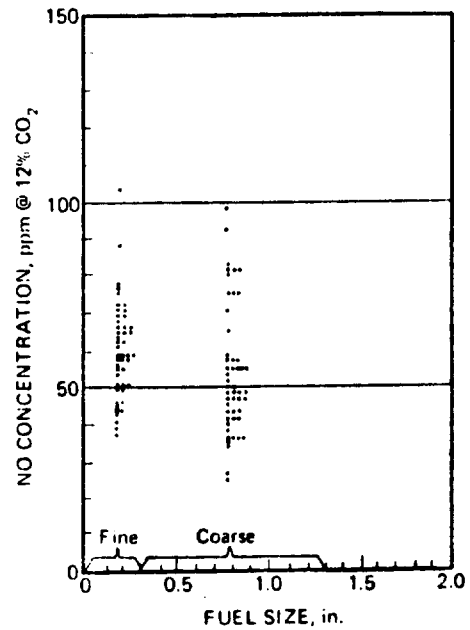
### 2. NO concentration as a function of fuel feed rate.



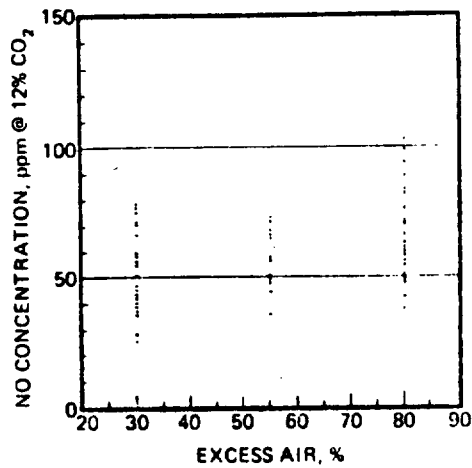
3. NO concentration as a function of combustion temperature.



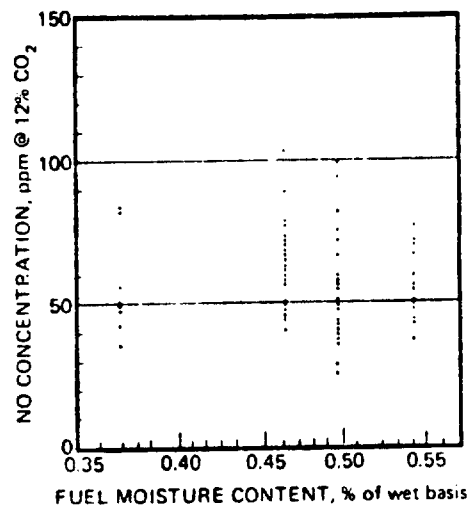
5. NO concentration as a function of fuel size.



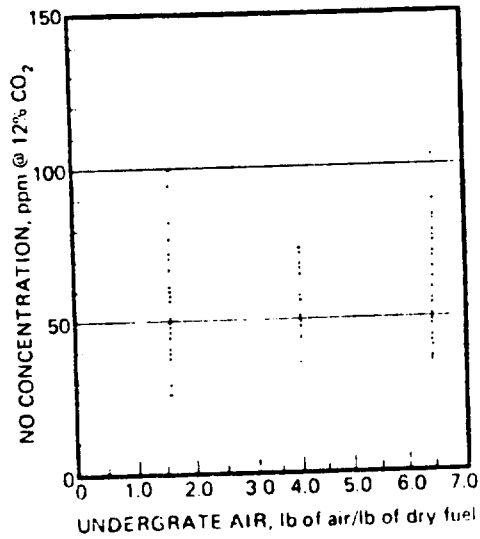
4. NO concentration as a function of excess air.



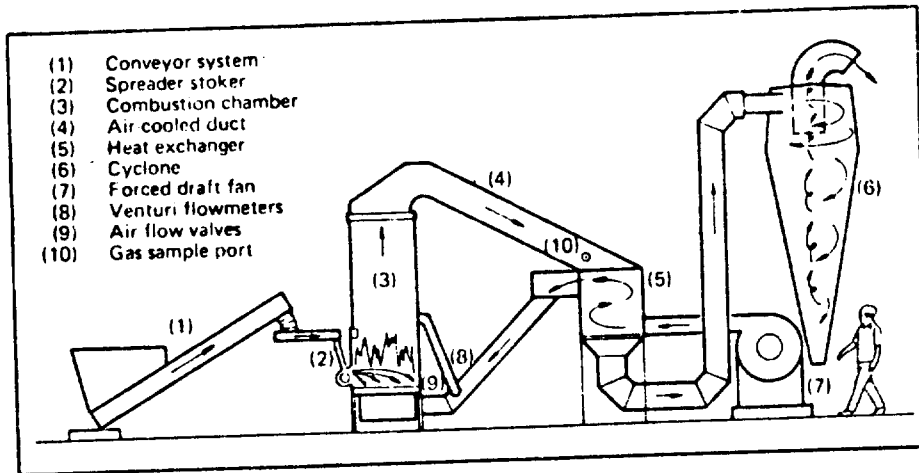
6. NO concentration as a function of fuel moisture content.



**7. NO concentration as a function of undergrate air.**



**8. Wood combustion test facility.**



## VITA

Richard Allen Kester, the son of Andrew Jackson and Roberta Mason Kester of Snohomish, Washington, was born May 11, 1942, in Everett, Washington. He attended the Snohomish public schools and graduated from high school in June 1960. In 1963, he graduated from Everett Junior College with an A.A. degree. He then entered the University of Washington and graduated with a B.S. degree in Civil Engineering in March of 1966. Mr. Kester then served 2 years with the U.S. Public Health Service, National Center for Air Pollution Control. He then spent over 10 years with the Puget Sound Air Pollution Control Agency before joining CH2M HILL. During this time, he entered graduate school at the University of Washington and earned an M.S. degree in Civil Engineering in March of 1972. In April 1975, he began his studies toward a Ph.D. in the Water and Air Resources Division, Department of Civil Engineering. On August 26, 1966, Mr. Kester married the former Diane Patricia Winter of Auburn, Washington. They have two children, Jason and Lisa.