



## *Project Summary*

# Effects of Fuel Properties and Atomization Parameters on NO<sub>x</sub> Control for Heavy Liquid Fuel Fired Package Boilers

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Experimental studies were conducted to (1) relate the formation of NO<sub>x</sub> in liquid fuel flames to the chemical/physical fuel properties and to the atomizer design, and (2) investigate the interaction between liquid sprays and airflow pattern for conditions typical of package boilers. These experiments were conducted in an 880 kW firetube boiler simulator and in a 21 kW tunnel furnace, under both normal and staged combustion conditions.

The fuels studied were primarily conventional heavy residual fuel oils covering a wide range of properties; however, shale- and coal-derived liquids were also tested. Fuel nitrogen content of the liquid fuels was found to be the only first order parameter affecting NO<sub>x</sub> emissions for a given spray/flow field configuration under excess air conditions. The tunnel furnace results indicated, however, that (during staged combustion) fuel nitrogen speciation has a second-order effect on the minimum achievable NO<sub>x</sub> level.

The boiler simulator studies showed that atomizer design, spray/flow field configuration, and fuel composition are significant dependent parameters affecting NO<sub>x</sub> emissions. Cold-flow spray characterization using laser diffraction showed that, in general, drop size information alone was insufficient to predict NO<sub>x</sub> emissions

in complex flow fields under normal combustion; however, staged combustion was found to be more effective with atomizers which produced smaller droplets.

Staged combustion was an effective NO<sub>x</sub> control for all liquid fuels tested. Optimization of staging parameters (e.g., mixing and primary zone time/temperature history) is indicated to achieve maximum NO<sub>x</sub> reduction using staged combustion.

*This Project Summary was developed by EPA's Industrial Environmental Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).*

### Introduction

Although increased coal use partially solves the U.S. energy crisis, many industrial users will continue to burn liquid fuels. However, the composition of these fuels will change as premium fuels are reserved for transportation and domestic use. Industrial users will be required to burn heavy petroleum-, coal-, and shale-derived residual oils, all of which have relatively high nitrogen contents and low hydrogen-to-carbon ratios. Unless appropriate pollution controls are applied, use of these fuels will increase NO<sub>x</sub> or particulate emissions from stationary combustors firing

these fuels. The production of  $\text{NO}_x$  and particulate in turbulent diffusion flames depends on the fuel composition and the fuel/air contacting process. The latter depends on the complex interaction of the liquid fuel spray from the combustion air flow field.

The overall goal of this study was to provide the information necessary to develop and generalize low  $\text{NO}_x$  oil burner technology for application to package firetube boilers. Specific objectives of the research were:

- To relate the formation of  $\text{NO}_x$  in liquid fuel flames to the chemical and physical fuel characteristics and to the atomizer design.
- To investigate the interaction between liquid sprays and the airflow pattern for conditions typical of package boilers under normal and staged conditions.

Two combustors (Figure 1) of different scales were used to provide information to assess the impact of liquid spray characteristics on  $\text{NO}_x$  formation and to relate the formation of  $\text{NO}_x$  under normal excess air and staged heat release conditions to the liquid fuel properties.

## Experimental Systems

### Tunnel Furnace

The down-fired tunnel furnace was designed to allow utilization of commercially available atomizers and to be fired with both artificial oxidants and air. The small-scale combustor was 2.1 m long with an I.D. of 0.2 m. The walls were insulating and high temperature castable refractory, and the nominal full-load heat release was 70,000 Btu/hr (21 kW). In certain investigations the combustion air was enriched or replaced with varying amounts of  $\text{CO}_2$ , Ar, and  $\text{O}_2$ , all of which were supplied from high pressure cylinders. The furnace was fired with a commercial air-assisted ultrasonic atomizer which was used because it provided adequate atomization of the viscous liquid fuels at these low flow rates.

### Boiler Simulator

Experiments at larger scale were carried out in an axisymmetric calorimetric combustor with a nominal firing rate of  $3 \times 10^6$  Btu/hr (880 kW). This pilot-scale combustor was divided into calorimetric sections cooled by heat transfer fluid and had a length of 3.2m and an I.D. of approximately 0.58m. The double-concentric burner could

accept a wide range of commercially available fuel nozzles. The unheated combustion air was supplied through an annular duct with an axial velocity of 30 m/sec. Swirl level could be varied by interchangeable fixed-vane annular swirl generators.

### Analytical System

Identical sampling and analysis systems, used for both combustors,

allowed for continuous monitoring of  $\text{NO}$ ,  $\text{NO}_x$ ,  $\text{CO}$ ,  $\text{CO}_2$ ,  $\text{O}_2$ , and  $\text{SO}_2$  using commercially available instruments. Flue gases were withdrawn from the appropriate combustor exhaust through a water-cooled stainless steel probe

### Fuel Effects

The work focused primarily on 13 petroleum-derived heavy fuel oils, but also considered three distillate oils, four

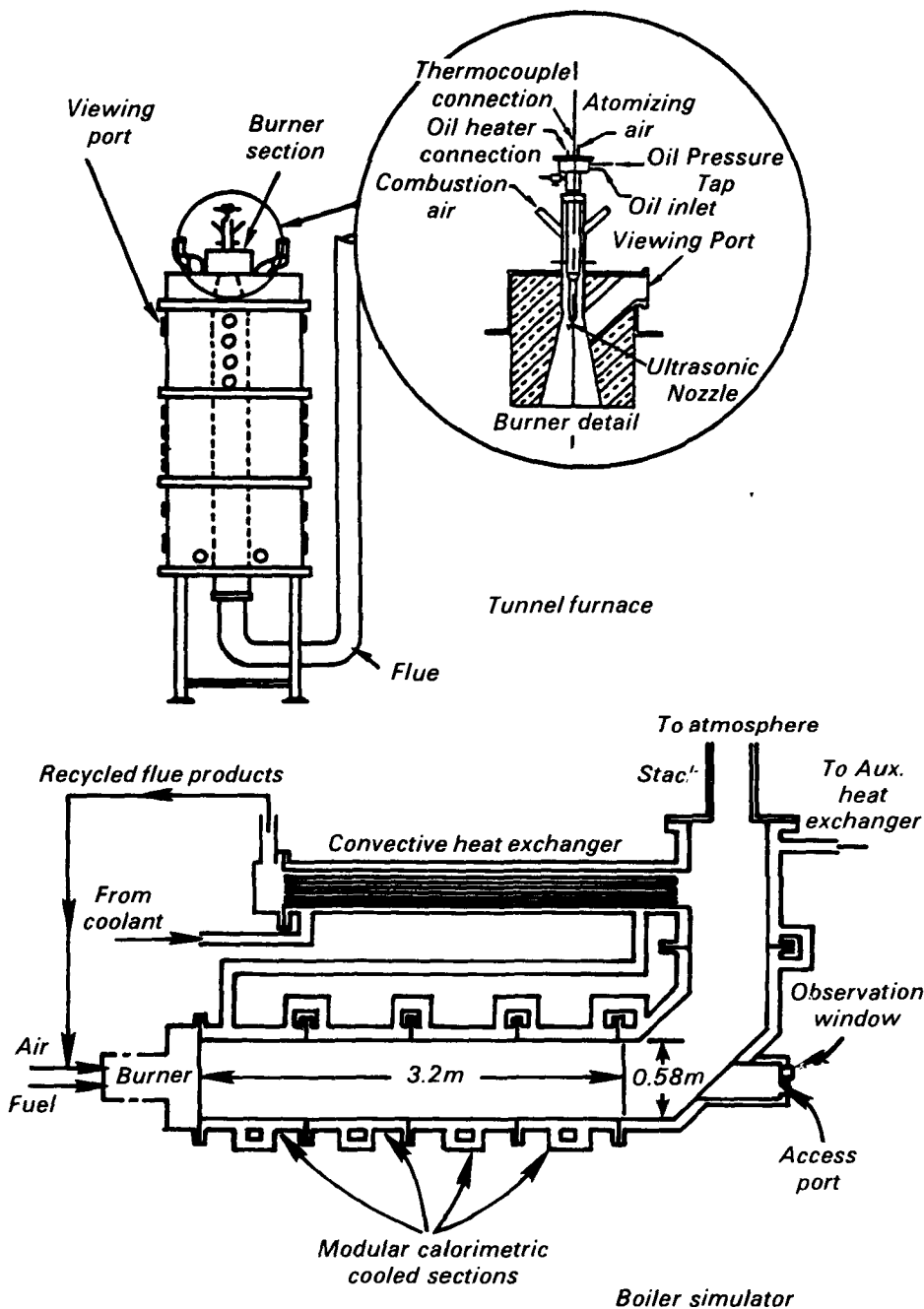


Figure 1. Combustors used in experiments.

shale oil blends, and one coal-derived liquid (Figure 2).

### Excess Air Combustion

Figure 3 is a composite plot of total and fuel NO<sub>x</sub> emissions from the tunnel furnace for a wide range of petroleum-, coal-, and shale-derived liquid fuels. Fuel NO<sub>x</sub> was defined by replacing combustion air with a synthetic oxidant mixture containing 21 percent O<sub>2</sub>, 20 percent CO<sub>2</sub>, and 59 percent Ar. CO<sub>2</sub> was added to the mixture to match theoretical flame temperature for the air and artificial oxidant cases. Both total and fuel NO<sub>x</sub> emissions increase with increasing fuel nitrogen content. These data are representative of a high mixing intensity burner with a finely atomized spray. Fuel NO<sub>x</sub> emissions increase by approximately 95 ppm per 0.1 percent nitrogen. Experiments were

also carried out with doped distillate oils: data from the doped distillates correlated well with those for the residual fuel oils which they simulated. Thus, it appears that, under excess air conditions with good fuel/air mixing, the only first order composition parameter involved in fuel nitrogen conversion is the total fuel nitrogen content of the fuel. This conclusion applies to petroleum-, coal-, and shale-derived liquid fuels. Data in Figure 4 for the boiler simulator furnace show a similar trend to those for the tunnel furnace. NO<sub>x</sub> emissions increase linearly with increasing fuel nitrogen content. However, the total emissions depend on the fuel/air contacting conditions illustrated by the differences shown for the two swirl levels. Emissions in the boiler simulator are the sum of those produced by the oxidation of molecular nitrogen (thermal NO) and those formed by the conversion

of fuel-bound nitrogen to NO (fuel NO). Extrapolation of the data in Figure 4 to zero percent nitrogen gives an estimate of the thermal NO contribution.

The use of synthetic oxidant, free from molecular nitrogen, allows the conversion of fuel nitrogen to fuel NO to be determined directly in the tunnel furnace. Figure 5 shows data for all the liquid fuels plotted in terms of percent conversion of fuel nitrogen to NO<sub>x</sub> as a function of fuel nitrogen content. In general, the percent conversion to NO<sub>x</sub> decreases as the fuel nitrogen increases. Alternate fuels follow much the same curve as the petroleum liquids, suggesting that the primary composition parameter is total fuel nitrogen, although there appear to be some second order fuel effects. The data in Figure 5 represent an upper level of fuel nitrogen conversion since they were obtained under well-mixed conditions.

#### Legend

- ◆ Alaskan diesel    ▽ California #1
- ▲ W. Texas diesel    ○ California #2
- California diesel    ◇ California #3
- Essex County    ⊠ California #4
- ▢ Middle east    ● Crude shale and blends with ⊠
- ⊠ Low sulfur #6
- △ Indo/Malaysian    ▽ Shale-derived DFM
- Desulfurized Venezuelan    ◆ SRC II
- ▢ Pennsylvania    ■ Synthoil blends with ●
- ◇ Gulf Coast
- Venezuelan    ● CH<sub>4</sub> + NH<sub>3</sub>
- Alaskan

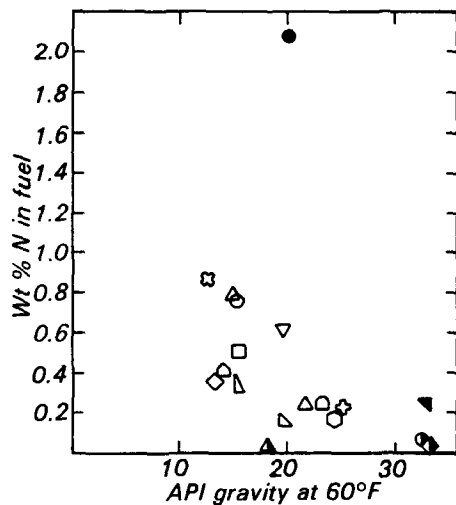


Figure 2. Fuel nitrogen versus API gravity.

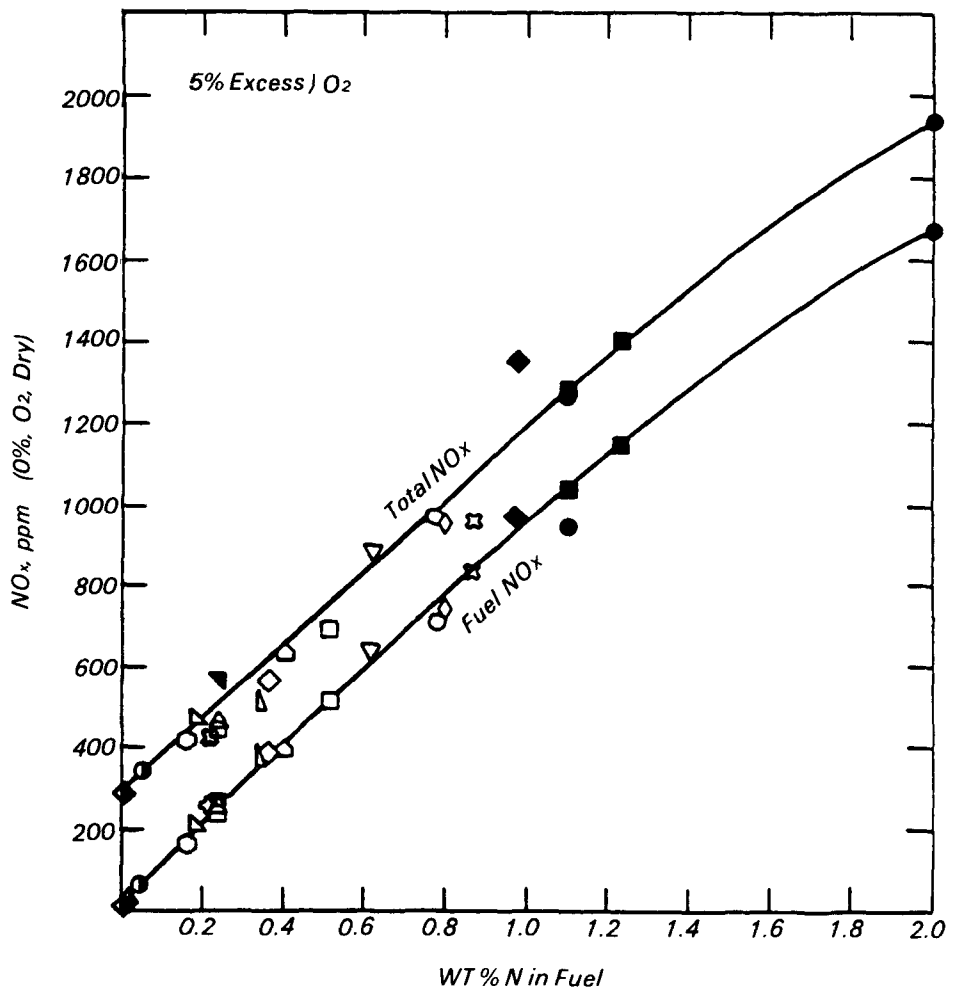


Figure 3. Effect of fuel nitrogen content on total and fuel NO<sub>x</sub> -- Tunnel Furnace.

## Staged Combustion

Staged combustion, a cost-effective control for the reduction of  $\text{NO}_x$ , involves operating the combustor to ensure that the fuel originally burns under oxygen-deficient conditions. Second stage air is then added to complete the oxidation process. Figure 6 shows data obtained from both the boiler simulator and the tunnel furnace

using the ultrasonic atomizer and operated at 3 percent overall excess oxygen. The data is presented as a function of the stoichiometric ratio of the primary zone, defined as the fraction of the theoretical air required for complete combustion. Since distillate oil is essentially nitrogen-free, these results represent the influence of the primary zone oxygen on  $\text{NO}_x$  produced from molecular nitrogen. The data for

the three residual oils indicate that, as the primary zone becomes progressively more fuel-rich, the ultimate  $\text{NO}_x$  emissions decrease dramatically to a minimum and then increase again. This upturn in emissions at very low stoichiometric ratios is probably due to an increase in the concentration of oxidizable fuel nitrogen species at the exit of the primary zone. Minimum  $\text{NO}_x$  emissions achievable under staged combustion conditions for a range of liquid fuels are compared in Figure 7. Minimum emissions from the boiler simulator are, in general, higher than those from the tunnel furnace, indicating that the small-scale furnace provides more optimum conditions for the minimization of  $\text{NO}_x$  production than does the boiler simulator; e.g., time and temperature in the fuel-rich zone. However, the general trends for both combustors are the same. Minimum  $\text{NO}_x$  emissions increase strongly with fuel nitrogen content up to fuel nitrogen levels of approximately 0.5 percent, and then show only a minor increase as the nitrogen content is increased above 2 percent by weight. The volatility of the nitrogen compounds in the liquid fuel appears to be the limiting factor in  $\text{NO}_x$  control by staged combustion. Experiments involving a residual fuel and a distillate oil doped with pyridine and thiophene to give the same nitrogen and sulfur content showed minimum emission levels under staged conditions 30 percent lower for the doped distillate than for the residual fuel.

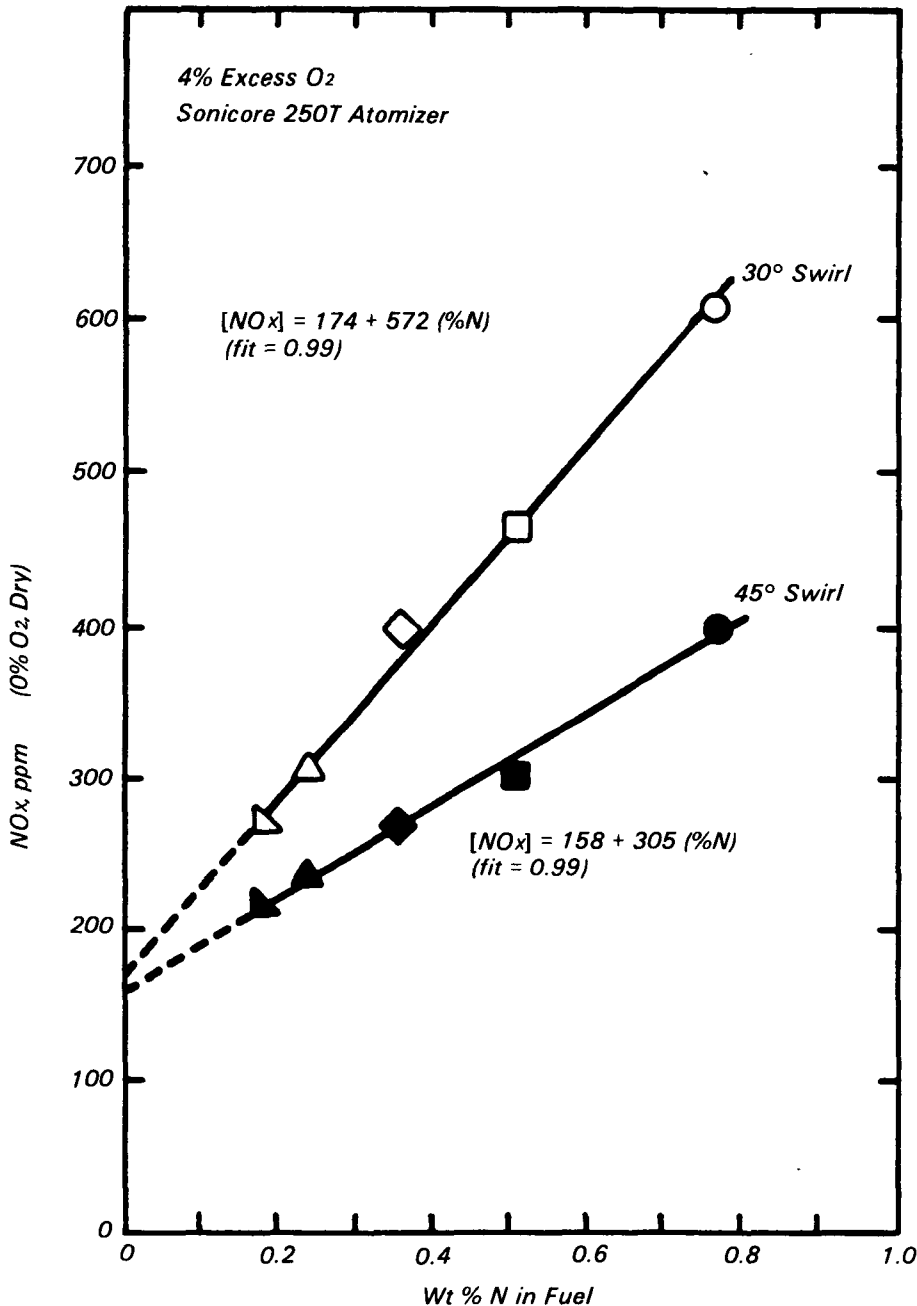


Figure 4. Effect of fuel nitrogen content -- boiler simulator.

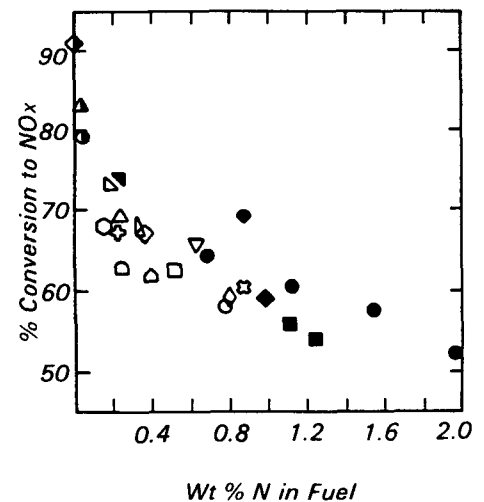


Figure 5. Composite fuel nitrogen conversion to  $\text{NO}_x$  as a function of fuel nitrogen content -- tunnel furnace.

## Atomizer Effects

A goal of this program was to assess the impact of atomizer characteristics on pollutant production under both excess air and staged combustion conditions. Commercial atomizers were selected for use in the boiler simulator and the characteristics of six of these atomizers were determined in a cold-flow test rig using a laser diffraction droplet size analyzer.

## Flow Field Interaction

In a given flow field,  $\text{NO}_x$  formation depends on the atomizer type. Figure 8 shows data for several commercial atomizers, four of which were characterized by the laser diffraction analyzer. The Peabody and Todd nozzles produced similar drop-size distributions (mean diameter approximately  $40 \mu\text{m}$ ). The largest mean drop size ( $90 \mu\text{m}$ ) was produced by the Monarch nozzle; the

Sonicore produced the smallest ( $21 \mu\text{m}$ ). Thus, emissions do not correlate directly with drop size. Figure 9 shows that the impact of nozzle design also depends on flow field characteristics. The effect of increasing swirl is seen to be different for each atomizer. Variations in swirl level produce a large change in  $\text{NO}_x$  emissions with the Monarch nozzle, and relatively mild change with the Delavan atomizer. The data in Figure 9b illustrate that drop size information alone is not sufficient to correlate emissions and that the flow field characteristics can have a significant effect with some atomizers.

## Staged Combustion

Increasing the mean drop size under staged combustion conditions effectively decreases the gas-phase residence time in the fuel-rich primary zone because it increases droplet lifetimes. Table 1 shows the effect of increasing mean drop size on staged  $\text{NO}_x$  emissions measured in the tunnel furnace for a given primary zone stoichiometry ratio and overall excess air level. As drop size increases, the final emission level also increases. However, this increase is less apparent for high nitrogen fuels. Figure 10 compares  $\text{NO}_x$  emissions obtained in the boiler simulator under staged combustion conditions with the Sonicore and the Monarch nozzles for two different fuels. In both instances the Sonicore nozzle gives higher  $\text{NO}_x$  emissions under excess air conditions, but lower emissions under staged combustion conditions. Smoke emissions increased dramatically as  $\text{NO}_x$  emissions decreased when burning petroleum-derived residual fuel, and nozzle design has the most significant impact on exhaust smoke number in this case. However, with shale-derived fuel, the smoke emissions are acceptable with both nozzles under all primary zone stoichiometries, but less  $\text{NO}_x$  is produced when using the nozzle with the smaller drop size distribution.

## Summary

The results of these small- and pilot-scale studies of the influence of fuel properties and spray/flow field interaction indicate that:

1. With liquid fuels, fuel nitrogen content is the primary fuel composition variable affecting fuel  $\text{NO}_x$  formation.  $\text{NO}_x$  emissions increase with increasing fuel nitrogen content for petroleum-, shale-, and coal-derived liquid fuels.

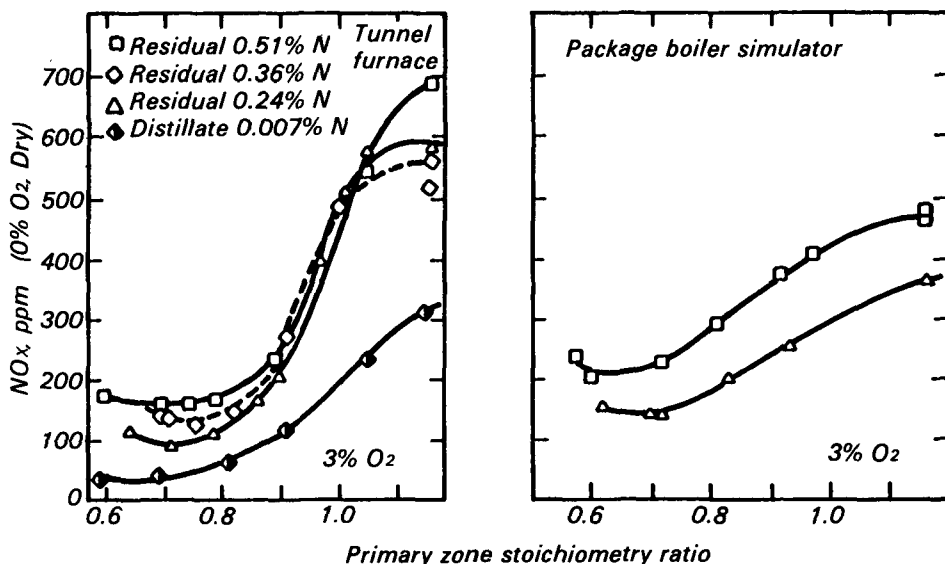


Figure 6. The effect of fuel composition on  $\text{NO}_x$  emissions under staged-combustion conditions.

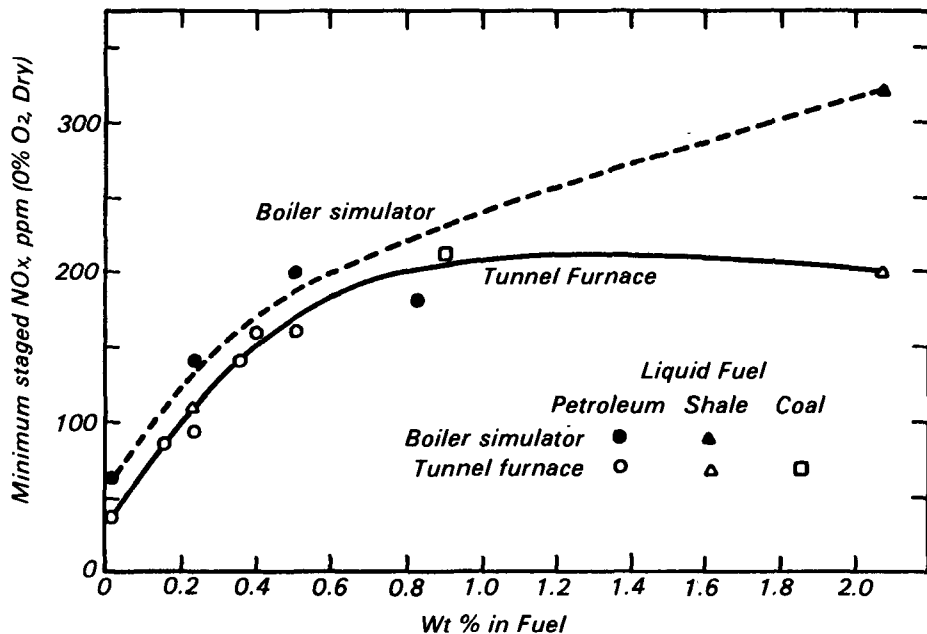


Figure 7. Minimum  $\text{NO}_x$  achievable under staged conditions.

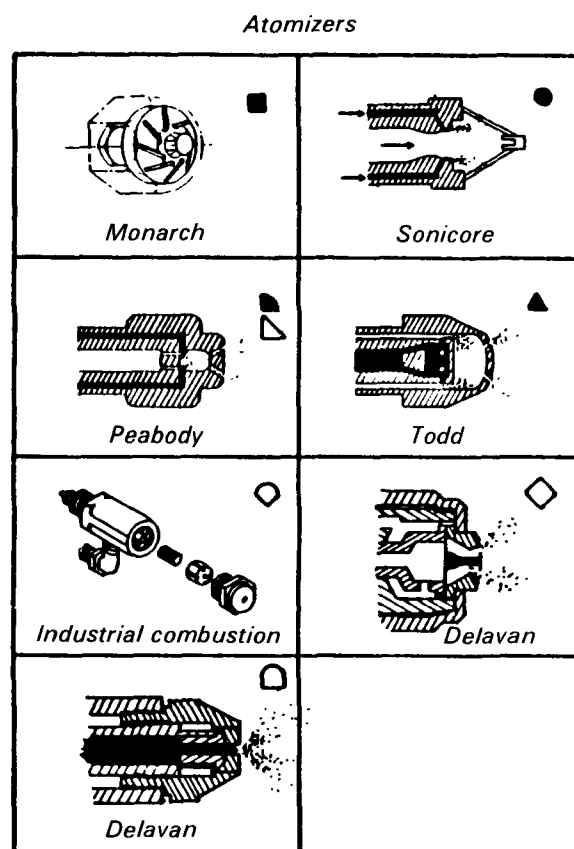
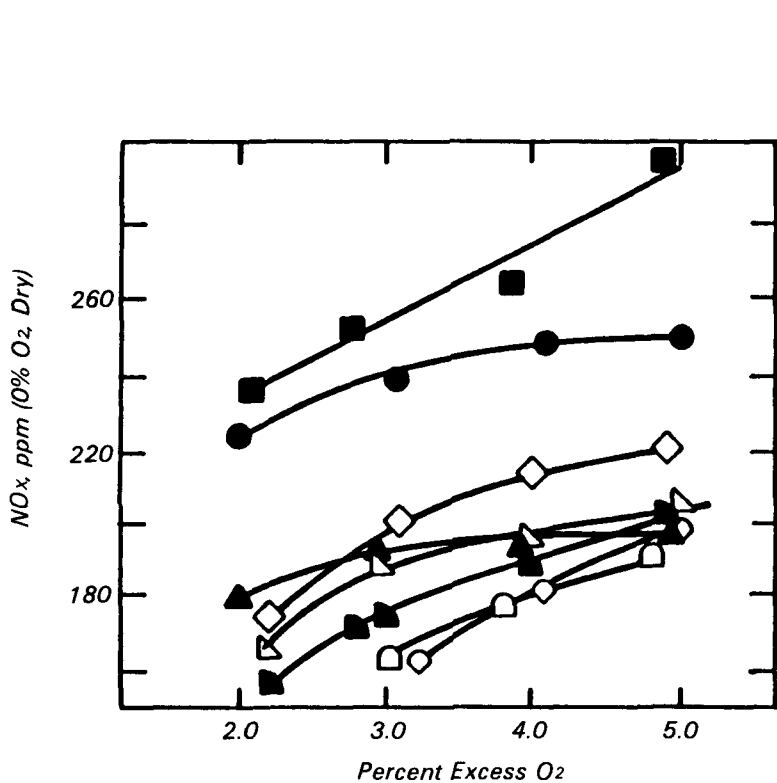
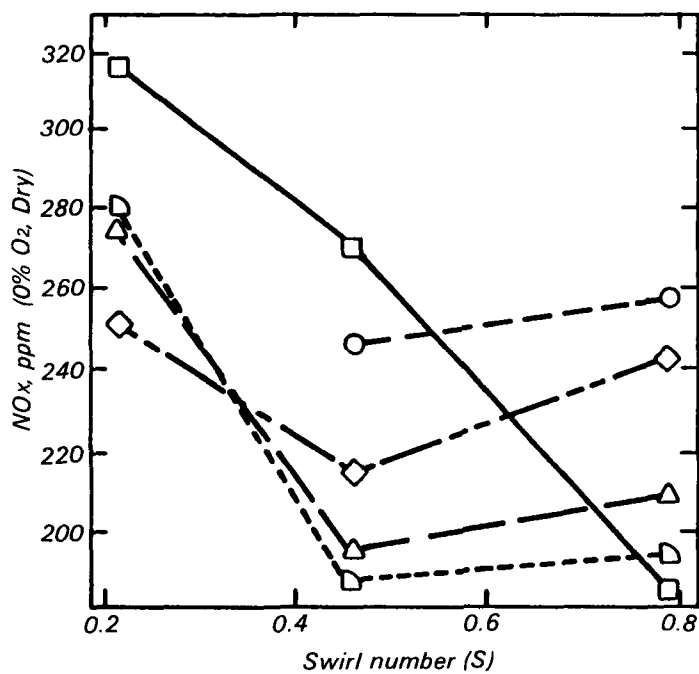
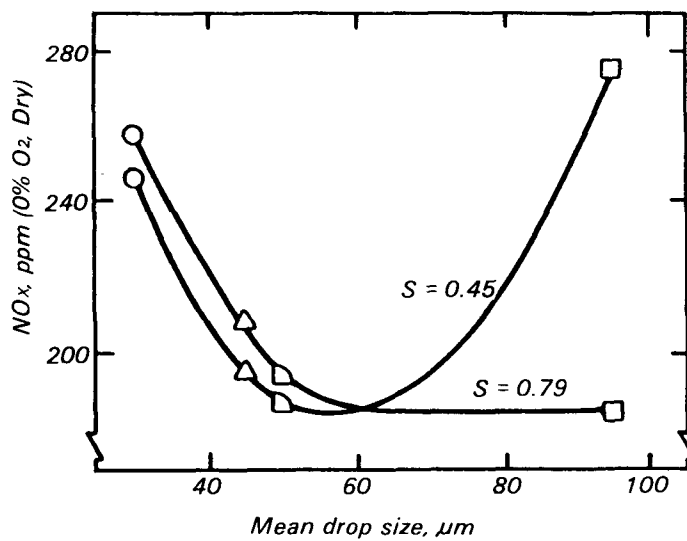
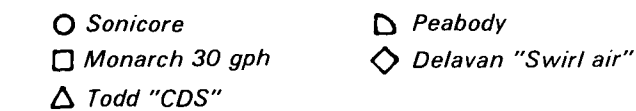


Figure 8. The effect of spray/flow field interactions.



(a)



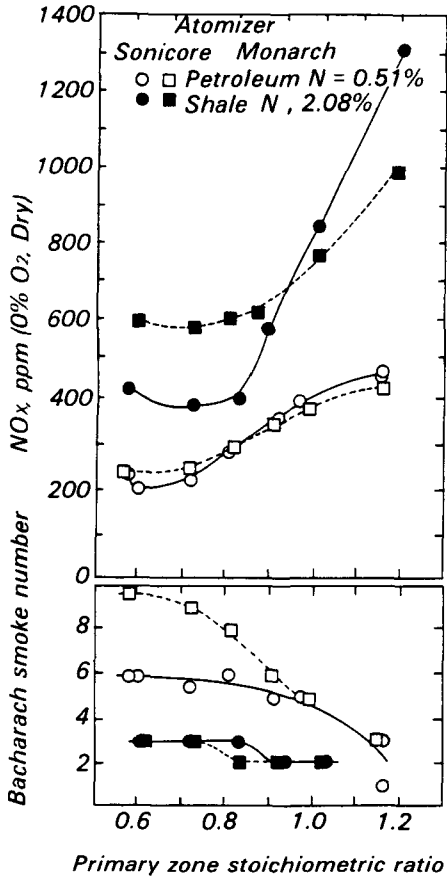
(b)

Figure 9. NO<sub>x</sub> emissions as a function of drop size.

**Table 1. Influence of Mean Drop Size on Staged NO<sub>x</sub> Emissions<sup>a</sup>**

Fuel Nitrogen, %	NO <sub>x</sub> ppm (0% O <sub>2</sub> , Dry)	
	20 μm	180 μm
0.007	48	75
0.16	85	105
0.24	110	140
0.51	165	170

<sup>a</sup>Tunnel furnace, primary zone stoichiometry is 0.70.



**Figure 10.** Influence of atomizer type on staged NO<sub>x</sub> and smoke emissions for the shale and petroleum fuels -- boiler simulation.

Staged combustion as a control technique is effective for all three fuels, and minimum levels achievable with high nitrogen alternate fuels approximate those of petroleum fuels with much lower nitrogen contents.

2. Decreasing droplet diameter reduces both NO<sub>x</sub> and smoke emissions under staged combustion conditions.
3. Fuel nitrogen speciation has an impact on the effectiveness of staged combustion as a control for minimizing NO<sub>x</sub> emissions produced with high nitrogen fuels.

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The complete report, entitled "Effects of Fuel Properties and Atomization Parameters on NO<sub>x</sub> Control for Heavy Liquid Fuel Fired Package Boilers," (Order No. PB 82-230 715; Cost: \$31.50, subject to change) will be available only from:

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