



## Project Summary

# NO<sub>x</sub> Abatement for Stationary Sources in Japan

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**Strict ambient air quality standards for SO<sub>2</sub> and NO<sub>x</sub> in Japan mandate the use of various air pollution technologies. This report is a compilation of information on the current status of NO<sub>x</sub> abatement technologies for stationary sources in Japan. The author obtained this information from electric power companies, various industries, and developers of numerous technology processes as well as from his own original research in the field. The report focuses on the Combustion Modification (CM) and Selective Catalytic Reduction (SCR) NO<sub>x</sub> abatement technologies. Information is provided on the development status, pilot and demonstration plant tests, technological problems, and costs associated with the use of these technologies in Japan. Detailed operation data are given to describe the commercial operation of SCR plants.**

***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).***

### **NO<sub>x</sub> Standards and Ambient Concentrations**

Recent air pollution control efforts in Japan have concentrated on NO<sub>x</sub> abatement, since ambient SO<sub>2</sub> concentrations already have been drastically reduced in response to stringent standards. In 1978, the ambient air quality standard for NO<sub>2</sub> was amended from 0.02 ppm to 0.04 - 0.06 ppm as a daily average. In regions with NO<sub>2</sub> concentrations above 0.06 ppm, the concentration will be reduced to 0.06 ppm by 1985. In regions with NO<sub>2</sub> concentrations of 0.04 - 0.06 ppm, efforts

will be made to keep the concentrations from substantially exceeding the present level. In areas with concentrations below 0.04 ppm, efforts must be made to maintain those levels. The new NO<sub>2</sub> standard is relaxed compared with the previous standard, 0.02 ppm as a daily average, but is still more stringent than the U.S. standard of 0.05 ppm as a yearly average.

In regions with large cities (such as Tokyo and Osaka), ambient NO<sub>2</sub> concentrations often exceed the standards, reaching 0.07-0.08 ppm as daily averages. The prefectural governments of Chiba, Kanagawa, and Mie have established even more stringent regulations and plan to reduce NO<sub>2</sub> concentrations to 0.04 ppm from the current 0.05-0.06 ppm. Even in regions with NO<sub>2</sub> concentrations below 0.04 ppm, NO<sub>x</sub> reduction is often required by local governments to prevent any further increase.

Nearly 2 million tons/yr of NO<sub>x</sub> are emitted in Japan, 60% from stationary sources and the rest from mobile sources. In large cities such as Tokyo and Osaka, about 60% of the NO<sub>x</sub> is emitted from mobile sources.

NO<sub>x</sub> emissions from gasoline-engine passenger cars manufactured since 1978 have been controlled by stringent regulations. The current limit is 0.25 g/km (8% of the NO<sub>x</sub> emissions from cars in 1973). NO<sub>x</sub> emissions from diesel-engine buses and trucks have been reduced to about 50% of the 1974 level. For stationary combustion sources, emission standards based on advanced combustion modification technology have been applied to reduce NO<sub>x</sub> by 30-70%.

With these efforts, ambient NO<sub>2</sub> concentrations are no longer increasing despite a continuing increase in the number of stationary and mobile sources. However, it is difficult to lower current NO<sub>2</sub> con-

centrations in large cities and industrial regions without more effective emission controls for diesel-engine cars and stationary sources.

## NO<sub>x</sub> Reduction for Stationary Sources

Nearly all NO<sub>x</sub> emissions are produced by the combustion of fossil fuels. In Japan, the major combustion fuel is heavy oil. This residue of the atmospheric distillation of crude oil has been used at the rate of nearly 200 million kl (kiloliters)/yr. Coal use decreased markedly between 1965 and 1975 and currently accounts for only 3% of Japan's total energy supply. However, coal consumption is expected to triple in the next 10 years. Imported LNG also accounts for about 3% of the energy currently used and is expected to nearly triple in 10 years.

Large stationary sources such as utility boilers have reduced NO<sub>x</sub> emissions 50-70% by applying combustion modifications (CM) including low excess air combustion, staged combustion, flue gas recirculation, and low-NO<sub>x</sub> burners. As a result, the NO<sub>x</sub> concentration in flue gas from utility boilers is minimal—150-300 ppm for coal, 80-120 ppm for oil, and 40-60 ppm for gas firing. Smaller boilers and furnaces have reduced NO<sub>x</sub> 30-50% by using low-NO<sub>x</sub> burners or by switching from heavy oil to kerosene.

For additional NO<sub>x</sub> abatement, several flue gas treatment (FGT) processes have been developed. Of all the processes, selective catalytic reduction (SCR), which uses ammonia and a catalyst at 300-400°C to control NO<sub>x</sub>, is presently the most advanced technology. Over 150 commercial SCR plants are in operation to remove 80-90% of the NO<sub>x</sub> emissions. Selective noncatalytic reduction (SNR), which uses ammonia at 800-1,000°C to remove 30-50% of NO<sub>x</sub> emissions, has been developed and applied to about 20 furnaces and industrial boilers. Wet and dry simultaneous SO<sub>x</sub> and NO<sub>x</sub> removal processes also have been developed but have not been applied commercially except for several small units.

SCR has been used most often for flue gas treatment because of its simplicity (which enables unattended operation), relatively high NO<sub>x</sub> removal efficiency (80-90%), and relatively low cost. Most of the new coal-fired utility boilers being planned will have SCR units. SCR will also be needed for some of the existing boilers even in regions with NO<sub>x</sub> concentrations below a 0.04 ppm daily average, due to local policies which forbid any increase in

NO<sub>x</sub> levels. For example, when a new boiler is installed at a power station, not only the new boiler, but also some of the existing boilers, will be required to have SCR units so that total NO<sub>x</sub> emissions from the station do not increase.

SCR is usually used with CM. For most boilers and furnaces, CM is applied first, followed by SCR in order to meet the stringent regulations. For over 90% NO<sub>x</sub> reduction, the combination of CM (to reduce 35-50% of the NO<sub>x</sub> emissions) and SCR (to remove 80-85% of the remaining NO<sub>x</sub>) is usually more economical than SCR by itself.

Typical examples of uncontrolled and controlled NO<sub>x</sub> concentrations in utility boiler flue gas are shown in Table 1. Examples of NO<sub>x</sub> regulations and emissions from utility boilers are shown in Table 2.

A new combustion process, in-furnace NO<sub>x</sub> removal, has been developed to remove about 50% of NO<sub>x</sub> by injecting a small portion of the fuel above the flame, followed by air addition to ensure complete combustion. Using this process, along with CM for utility boilers, NO<sub>x</sub> may be reduced to 100 ppm for coal, 50 ppm for oil, and 20 ppm for gas.

Fluidized-bed combustion, gasification, and liquefaction of coal all have been tested in Japan, but these technologies are not as advanced there as they are in the U.S. This is because Japan must import coal and because these technologies are unable to meet the stringent Japanese NO<sub>x</sub> emission regulations. Most of the

new coal-fired boilers in Japan will use conventional pulverized coal combustion with CM, SCR, and FGD.

## SCR and SCR/FGD Characteristics

SCR problems of the past have been solved by recent improvements. The major problems were: 1) poisoning of the catalyst by SO<sub>x</sub> in the gas, 2) dust plugging of the catalyst, and 3) deposition of ammonium bisulfate in the air preheater downstream of the SCR reactor. Catalyst poisoning has been eliminated by using catalysts based on TiO<sub>2</sub> instead of Al<sub>2</sub>O<sub>3</sub> or Fe<sub>2</sub>O<sub>3</sub>. The use of parallel-flow honeycomb, tube, or plate catalysts or a parallel passage reactor eliminates dust plugging. Ammonium deposits can be prevented by maintaining the concentration of unreacted ammonia in the reactor outlet gas below 5 ppm and using a low-oxidation catalyst. To do this, 0.82-0.95 mole NH<sub>3</sub> is usually used per mole NO<sub>x</sub> to obtain 80-90% NO<sub>x</sub> removal with less than 5 ppm unreacted NH<sub>3</sub>, while SO<sub>2</sub> oxidation is kept below 1%.

SCR and FGD system applications to boilers and other gas sources are shown schematically in Figure 1. SCR is easily applied to boiler economizer outlet gas at 300-400°C as shown in the A portion of the figure. For SO<sub>x</sub>-rich gases, FGD may be applied downstream of SCR as shown in B. At an early stage of development, SCR was applied downstream of FGD systems, as shown in C, to protect the catalyst from SO<sub>x</sub> attack. System C, however, is expensive; it requires large amounts

**Table 1.** Examples of Controlled and Uncontrolled NO<sub>x</sub> Concentrations in Utility Boiler Flue Gas

Fuel	Outlet NO <sub>x</sub> Concentration, ppm			Percent Control		Total % Control
	Without Control, ppm	Controlled by CM	Controlled by CM and SCR	by CM	by SCR	
Gas	200	50	10	70	83	95
Oil	300	100	20	67	80	93
Coal	600	250	50	58	80	91

**Table 2.** Examples of NO<sub>x</sub> Regulations and Emissions from Utility Boilers, ppm

Fuel	Boiler	Central Government	Local Agreement	Actual Emission
Gas	Existing	100	60	60 <sup>a</sup>
Gas	New	60	10	8 <sup>b</sup>
Oil	Existing	150	100	100 <sup>a</sup>
Oil	New	130	25	20 <sup>b</sup>
Coal	Existing	400	159 <sup>c</sup>	170 <sup>a</sup>
Coal	New	400	170	160 <sup>d</sup>

<sup>a</sup>By combustion modification (CM).

<sup>b</sup>By CM and selective catalytic reduction (SCR).

<sup>c</sup>Desired by local government.

<sup>d</sup>By CM and partial SCR.

of energy for gas reheating. System B has become popular as SO<sub>x</sub>-resistant catalysts have been developed.

System D is often used for flue gas from a low-sulfur coal. In this system, the boiler economizer outlet gas is first treated by a hot electrostatic precipitator (ESP) and then by SCR and FGD. A cold ESP is not highly efficient for flue gas from low-sulfur coal. For high- and medium-sulfur coals, system B is preferable. System B may also be useful for low-sulfur coal if both the cold ESP and the FGD unit are designed for sufficient dust removal.

Low-temperature catalysts have been used for 200-250°C gases such as that produced by coke ovens, as shown in system E of Figure 1. Since ammonium bisulfate deposits on the catalyst at these low temperatures, the catalyst requires occasional heating to 400°C to remove the bisulfate.

When wet FGD is applied downstream of SCR or SNR, the ammonia present in the reactor outlet is caught by the FGD system and goes into the wastewater. In some cases it may be necessary to use the activated sludge process to remove ammonia from the wastewater.

### Cost of NO<sub>x</sub> Abatement for Stationary Sources

The investment cost for a combustion modification system is shown in Table 3. Costs range from ¥400\* to 800/ Nm<sup>3</sup> of flue gas or ¥1,200-2,400/kW for 55-70% reduction using a combined low-NO<sub>x</sub> burner, staged combustion, and flue-gas recirculation system.

SCR costs for new 700 MW gas-, oil-, and coal-fired utility boilers are shown in Table 4. For cost estimation purposes, it was assumed that flue gases leaving the boiler economizer at 330-400°C are treated in two equal-size reactors in parallel and that unreacted NH<sub>3</sub> is kept below 5 ppm. The investment cost for 80% NO<sub>x</sub> removal is about ¥2,500/kW for gas, ¥4,100-6,200/kW for oil and ¥6,700-8,400/kW for coal. The cost differences are due to the varying amounts of catalyst required. For example, a small amount of a very active catalyst is used for gas streams while a larger amount of a less active catalyst which is resistant to SO<sub>x</sub> and dust erosion is used for dirty flue gas (oil or coal streams). Compared with 80% removal, 90% removal costs 15% more for gas, 25-30% more for oil, and 30% more for coal. The investment cost of an SCR system for an existing boiler is 10-50% more than for a new boiler.

(\* ) \$1 = ¥250.

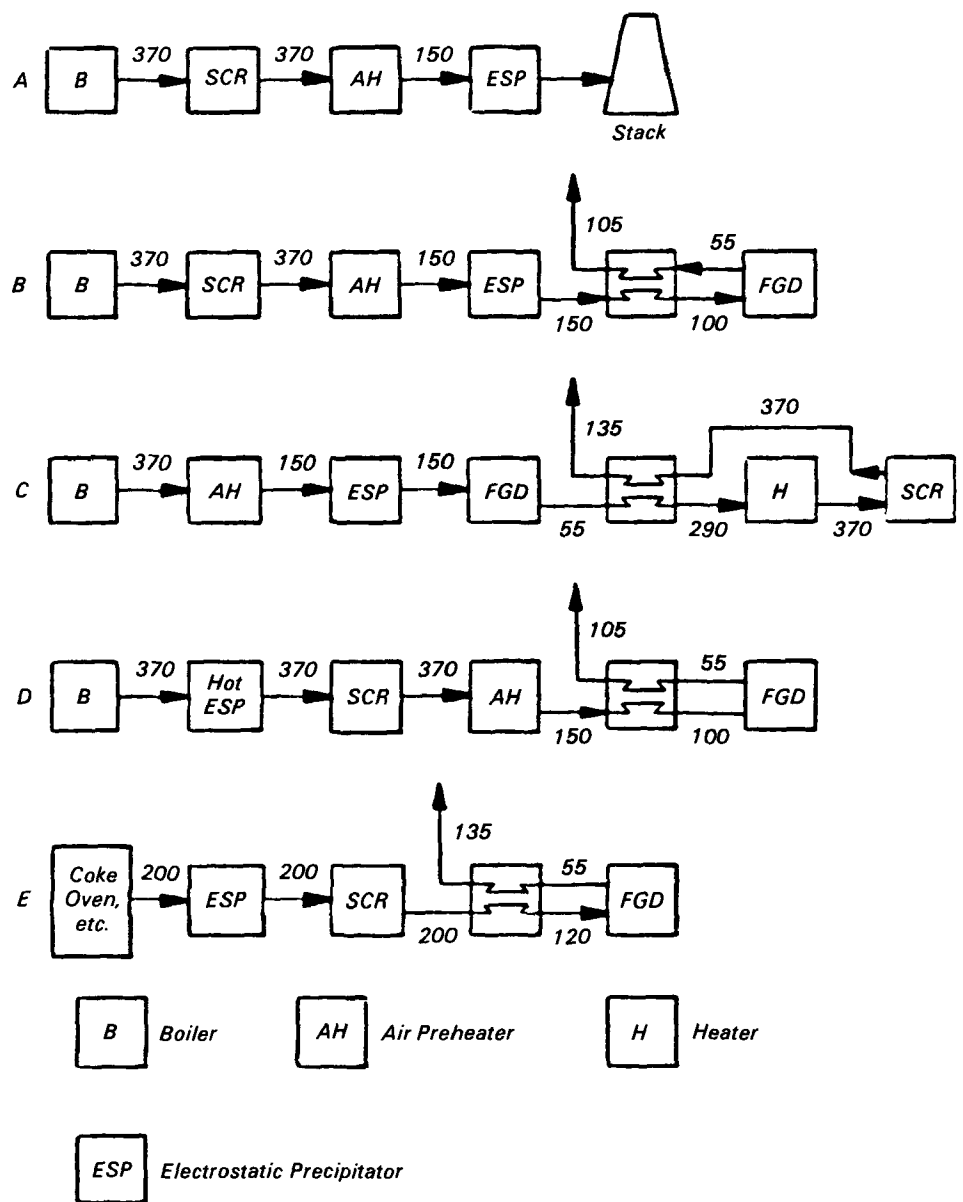


Figure 1. SCR and FGD system arrangements in use in Japan. (Numbers indicate gas temperature in °C.)

Table 3. CM Investment Cost

Method	NO <sub>x</sub> removal, %	¥/Nm <sup>3</sup>	¥/kW	\$/kW <sup>a</sup>
Low-NO <sub>x</sub> Burner	20-40	100-200	300-600	1.2-2.4
Combined System <sup>b</sup>	50-70	400-800	1200-2400	4.8-9.6

<sup>a</sup>\$1 = ¥250.

<sup>b</sup>Low-NO<sub>x</sub> burner, staged combustion, and flue gas recirculation.

The dirtier the gas, the shorter the life of the catalyst. Therefore, the annualized SCR cost/kWhr is higher when SCR is used with dirty gas. On the other hand, the annualized cost per pound of NO<sub>x</sub> re-

moved is lower with NO<sub>x</sub>-rich gas. The cost per pound for 80% removal is 10-17 percent lower than the cost for 90% removal. NO<sub>x</sub> removal of 90% with a low level of unreacted NH<sub>3</sub> (about 5 ppm) is

**Table 4. SCR Cost for 700 MW New Boiler (1981 Cost) (70% Boiler Utilization 4,292,000 MWhr/yr)**

Fuel	Gas		Low-S Oil		High-S Oil		Low-S Coal		High-S Coal		High-S Coal	
Inlet NO <sub>x</sub> , ppm	60		100		200		300		300		600	
Catalyst Type	Pellet		Honeycomb		Honeycomb		Honeycomb		Honeycomb		Honeycomb	
Cost, 10 <sup>6</sup> ¥/m <sup>3</sup>	3.0		3.3		3.3		3.5		3.5		3.5	
Life, years	4		3		3		2		2		2	
NO <sub>x</sub> Removal, percent	80	90	80	90	80	90	80	90	80	90	80	90
Investment Cost <sup>a</sup> , 1,000 ¥/kW	2.47	2.80	4.13	5.11	6.23	7.93	6.69	8.51	7.26	9.10	8.44	10.76
Annualized cost <sup>b</sup> , ¥/kWhr	0.17	0.20	0.28	0.35	0.44	0.56	0.59	0.76	0.65	0.82	0.81	1.04

<sup>a</sup>Including initial charge of catalyst, civil engineering, and test operation.

<sup>b</sup>Including 10% interest and 7 years depreciation.

not easy to obtain with a large amount of gas from a utility boiler, because both gas velocity and NO<sub>x</sub> concentrations vary across the duct at the reactor inlet.

The investment and annualized SCR costs for 80% SO<sub>2</sub> removal for coal-fired boilers are about one-third of those for 90% SO<sub>2</sub> removal using the wet lime/limestone FGD process. On the other hand, SCR is more expensive than CM. Although the investment cost of SCR for a gas-fired boiler is similar to that of CM in the combined system (Table 3), the annualized cost of SCR may be considerably higher than CM, which has low operating costs. Therefore, for NO<sub>x</sub> abatement, CM should be used first and SCR should be used in combination with CM when CM alone is not sufficient to meet control regulations. One CM technique, flue gas recirculation, is relatively expensive and is not highly efficient for coal. For this reason, flue gas recirculation may not be useful when SCR is applied to coal-fired boilers.

The costs of other FGT processes are uncertain because they have not been used widely in continuous commercial

operation. However, experience with Thermal DeNO<sub>x</sub>, a type of SNR used with an oil-fired utility boiler, indicates that its cost is about half that of SCR although the NO<sub>x</sub> removal efficiency is also half as much (40% versus 80%).

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*The complete report, entitled "NO<sub>x</sub> Abatement for Stationary Sources in Japan," (Order No. PB 83-207 639; Cost: \$37.00, subject to change) will be available only from:*

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