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Work Order No. 05-06

**ASSESSMENT OF NO_x EMISSIONS REDUCTION STRATEGIES FOR
CEMENT KILNS - ELLIS COUNTY AND ACROSS TEXAS**

PRELIMINARY PHASE I DRAFT REPORT

TCEQ Contract No. 582-04-65589
Work Order No.05-06

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1.0 INTRODUCTION

This study was conducted to explore potential NO_x emissions reduction strategies for cement kilns in Ellis County. This project included assessing existing NO_x control technologies as well as new technologies that have not been previously considered by TCEQ. This project evaluated the general performance and cost of applicable NO_x control technologies for the cement kilns present in Ellis County and further evaluated the application of these control technologies to the site-specific requirements of the kilns and raw materials present in Ellis County.

This study was initiated in order to fulfill obligations in the Dallas/Fort Worth Litigation Settlement Agreement. TCEQ, in consultation with the Plaintiffs, EPA, and Intervenor the Portland Cement Association, developed the scope of work and selected Eastern Research Group (ERG) and a panel of experts (the ERG Team) to perform this cement kiln NO_x control technologies study. This study evaluated the applicability of existing control technologies and the potential availability of new air pollution control technologies for cement kilns in the Dallas/Ft Worth ozone nonattainment area. This report evaluates and establishes type of controls that may be technically and economically applied to the three nonattainment area cement plants with specific evaluations of applicability to the 10 cement kilns located at the three plants.

The intent of this study is to establish impartial parameters for the determination of applicable controls. Controls are included to the extent that they were technically feasible. Control costs and cost effectiveness values, in terms of cost per ton of NO_x controlled, are then assigned to all technically feasible controls. The ERG Team has not applied value judgments or assessed the economic viability of technically feasible controls. The ERG Team has not excluded control technology on the basis of cost. This study puts forward technically viable NO_x control technologies and evaluates their cost effectiveness for specific application to Ellis County cement kilns with the intent of allowing TCEQ and other interested parties to have sufficient information make control technology selections.

USEPA published a review of NO_x controls for the cement industry titled “NO_x Control Technologies for the Cement Industry”, U.S. Environmental Protection Agency Office of Air Quality Planning and Standards, September 19, 2000 (EPA 2000). Review of this document indicated that it was a comprehensive review of the combustion and process modification

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techniques that are currently in use in the US cement industry. This report was determined to be a useful baseline and point of departure for this study. The ERG Team has reviewed and edited this report to update, make consistent with the ERG Teams understanding of processes and controls and make relevant to Texas and Ellis County plants and conditions. Sections 2 and 3 of this report are largely based on the EPA 2000 report. Section 4 of this report focuses on three control technologies that go beyond the combustion and process modifications that are widely in use in the US. The ERG team viewed research of these three technologies and their potential for application to Ellis County kilns as the essential point of departure from the EPA 2000 Report and as the key developments in prospective control options that have occurred since EPA's Report was written. The three developments in technology are: more widespread application of selective non-catalytic reduction (SNCR), a European application of selective catalytic reduction (SCR) and the use of low temperature oxidation in refineries and boilers making this technology worthy of evaluation as a transferable technology for cement kilns.

Attachment 1 to this report is the ERG Team's summary of the technology evaluations contained in the EPA 2000 Report (Table 1). As stated above, Table 1 does not represent the ERG Teams conclusions about control technologies, it represents a baseline or point of departure for this study. Many of the combustion and process modifications for controlling NO_x presented in this report are already in place in the Ellis County kilns and are well understood by the industry and the regulatory community. This report will analyze and update the information on combustion and process controls, but will also focus on the potential application of the three active control technologies: SNCR, SCR and low temperature oxidation.

2.0 DEFINING PARAMETERS FOR CONTROL EVALUATION

2.1 Overview of Portland Cement Manufacturing Process and NO_x Generation

Portland cement, a fine gray or white powder, is the generic term for the type of cement used in virtually all concrete, which is a mixture of aggregates (e.g., crushed stone, gravel, sand), water, and cement. The American Society for Testing and Materials (ASTM) defines portland cement as "hydraulic cement (cement that not only hardens by reacting with water but also forms a water-resistant product) produced by pulverizing clinkers consisting essentially of hydraulic

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calcium silicates, usually containing one or more of the forms of calcium sulfate as an inter ground addition."

In 2004, the portland cement industry in the U.S. consisted of 114 facilities in 37 States (and 2 facilities in Puerto Rico). Approximately 90 million tons of portland cement were produced in 2004, with California, Texas, Pennsylvania, Michigan, Missouri, and Alabama accounting for approximately 50 percent of the production [USGS].

Hydraulic portland cement, the primary product of the cement industry, is made from clinker blended with gypsum. Clinker is produced by heating a mixture of limestone, clay, and other ingredients to incipient fusion at a high temperature. Limestone is the single largest ingredient required in the cement-making process, and most cement plants are located near large limestone deposits. Portland cement is used in almost all construction applications including homes, public buildings, roads, industrial plants, dams, bridges, and many other structures.

In the cement-making process, the solid raw materials are heated to their clinkering temperature, typically 1400 to 1500 °C (2550 to 2750 °F), by burning various fuels such as coal. Portland cement has been defined as "a hydraulic cement produced by pulverizing portland cement clinker and usually containing calcium sulfate."¹ Portland-cement clinker has been defined as "a clinker, partially fused by pyroprocessing, consisting predominantly of crystalline hydraulic calcium silicates."¹ Burning an appropriately proportioned mixture of raw materials at a suitable temperature produces hard fused nodules called clinker, which are further ground to a desired fineness.

2.1.1 Description of the Cement Industry

About 77.6 million metric tons of gray portland and 274,000 metric tons of white cement were produced in a total of 198 cement kilns at 118 plants in the United States in 1998.⁵ This was a 6.0 percent increase from the 1990 reported total production of 73.5 million metric tons. Cement industry annual clinker capacity steadily declined from the 1975 peak through 1990 and has steadily increased since the 1990 low. While the number of kilns has dropped off sharply, average kiln size has increased. Since 1973 when average kiln size was 173,000 metric tons, average kiln size has now reached 393,000 metric tons. Although 42 cement companies produced clinker in 1998, the top 5 companies provided about 44.2 percent of the total finish grinding capacity. This is evidence of a high concentration of the U.S. production among a

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limited number of top producers. California and Texas are the two largest states in terms of clinker capacity with Pennsylvania, Missouri, and Alabama rounding out the top five. Fourteen states and the District of Columbia had no cement clinker-producing plants in 1998.⁵

The large majority of the cement plants (about 82.4 percent) in the United States are coal fired with about 2.8 percent using natural gas, and 0.9 percent using oil as the primary fuel.⁵ The remaining 13.9 percent of the plants used other combinations, e.g. coal/waste as primary fuel. In 1998, 11 plants used waste as a primary fuel with 49 plants reporting waste as an alternate fuel.

2.1.2 Overview of Cement Manufacturing Process

The process of portland cement manufacture consists of:⁶

- Quarrying and crushing the rock,
- Grinding the carefully proportioned materials to high fineness,
- Subjecting the raw mix to pyroprocessing in a rotary kiln, and
- Grinding the resulting clinker to a fine powder.

The choice between the wet or dry process for cement manufacturing often depends upon the moisture content in the raw feed materials mined from quarries. If the moisture content of the feed materials is already very high (15 to 20 percent), a wet process may be attractive. The recent trend, however, has been toward the dry process with preheater/precalciner systems. In 1998, about 20.6 million metric tons of clinker were produced by the wet process with 57.4 million metric tons produced by a dry process. Within the dry process category, 14.2 million metric tons were produced by facilities equipped with a preheater system and 26.1 million metric tons were produced by facilities equipped with a precalciner system.⁵

The different steps involved in the cement manufacturing process are described in the following subsections.

2.1.2.1 Raw Materials and Kiln Feed Preparation

The bulk of raw materials originates in the plant quarry. A primary jaw or roll crusher is frequently located within the quarry and reduces the quarried limestone or shale to about 100 mm top size. A secondary crusher, usually roll or hammer mills, typically gives a product of about 10 to 25 mm top size. Combination crusher-dryers can utilize exit gases from the kiln or

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clinker cooler to dry wet material during crushing. Each of the raw materials is stored separately and proportioned into the grinding mills separately using weigh feeders or volumetric measurements. Ball mills are used for both wet and dry processes to grind the material to a fineness such that only 15 to 30 wt% is retained on a 74- μm (200 mesh) sieve.

In the wet process the raw materials are ground with about 30 to 40 percent water, producing a well-homogenized mixture called slurry. Raw material for dry process plants is ground in closed-circuit ball mills with air separators, which may be adjusted for the desired fineness. Drying may be carried out in separate units, but most often is accomplished in the raw mill simultaneously with grinding. Waste heat can be utilized directly in the mill by coupling the raw mill to the kiln- and/or clinker cooler exhaust. For suspension preheater-type kilns, a roller mill utilizes the exit gas from the preheater to dry the material in suspension in the mill. A blending system provides the kiln with a homogeneous raw feed. In the wet process the mill slurry is blended in a series of continuously agitated tanks in which the composition is adjusted as required. These tanks may also serve as kiln feed tanks or the slurry may be pumped to large kiln feed basins. Dry process blending is usually accomplished in a silo with compressed air.⁶

2.1.2.2 *Pyroprocessing*

All cement clinker is produced in large rotary kiln systems. The rotary kiln is a refractory brick-lined cylindrical steel shell [3 to 8 m (10 to 25 ft) diameter, 50 to 230 m (150 to 750 ft) long] equipped with an electrical drive to rotate the kiln on its longitudinal axis at 1 to 3 rpm. It is a countercurrent heating device slightly inclined to the horizontal, so that material fed into the upper end travels slowly by gravity to be discharged into the clinker cooler at the lower, discharge end. The burners at the firing end, i.e., the lower or discharge end, produce a current of hot gases that heats the clinker, and the calcined and raw materials in succession, as it passes upward toward the feed end. Refractory bricks of magnesia, alumina, or chrome-magnesite combinations line the firing end. In the less heat-intensive midsection of the kiln, bricks of lower thermal conductivity are often used. Abrasion-resistant bricks or monolithic castable linings are used at the feed end.⁶

Pyroprocessing may be conveniently divided into four stages, as a function of location and temperature of the materials in the rotary kiln.⁸

1. Evaporation of uncombined water from raw materials, as material temperature

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increases to 100°C (212°F);

2. Dehydration, as the material temperature increases from 100°C to approximately 430°C (800°F) to form dehydrated clay minerals composed of oxides of silicon, aluminum, and iron;
3. Calcination, during which carbon dioxide (CO₂) and CaO are formed from calcium carbonates, primarily between 900°C (1,650°F) and 982°C (1,800 °F); and
4. Reaction, of the oxides in the burning zone of the rotary kiln, to form cement clinker at temperatures of approximately 1,510°C (2,750°F).

The duration and location of these stages in an actual kiln depend upon the type of process used, e.g., wet or dry, and the use of preheaters and precalciners as discussed in the following section.

It is desirable to cool the clinker rapidly as it leaves the burning zone. Heat recovery, preheating of combustion air, and fast clinker cooling are achieved by clinker coolers of the reciprocating-grate, planetary, rotary, or shaft type. Most commonly used are grate coolers where the clinker is conveyed along the grate and subjected to cooling by ambient air, which passes through the clinker bed in cross-current heat exchange. The air is moved by a series of undergrate fans and becomes preheated to 370 to 1,000°C (700 to 1,830°F) at the hot end of cooler. A portion of the heated air serves as secondary combustion air in the kiln. Primary air is that portion of the combustion air needed to carry the fuel into the kiln and disperse the fuel.⁶

2.1.3 Kiln Types and Operation

There are four main types of kilns used in portland cement manufacture:

- long wet kilns,
- long dry kilns,
- dry kilns with preheaters, and
- dry kilns with precalciners.

Wet kilns tend to be older units and are often located where the moisture content of feed materials from local sources (quarries) tends to be high. In Ellis County, only wet process and dry preheat/precalcination process kilns are used.

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2.1.3.1 Long Wet Kilns

In a long wet-process kiln, the slurry introduced into the feed end first undergoes simultaneous heating and drying. The refractory lining is alternately heated by the gases when exposed and cooled by the slurry when immersed; thus, the lining serves to transfer heat as do the gases themselves. Large quantities of water must be evaporated; thus most wet kilns are equipped with chains suspended across the cross section of the kiln to maximize heat transfer from the gases to the slurry, or chain garlands that serve to recuperate heat and simultaneously convey the slurry. After most of the moisture has been evaporated, the nodules, which still contain combined water, move down the kiln and are gradually heated to about 550°C (1,022°F) where the calcination reactions commence. The calcined material further undergoes clinkering reactions. As the charge leaves the burning zone and begins to cool, clinker minerals crystallize from the melt, and the liquid phase solidifies. The granular clinker material drops into the clinker cooler for further cooling by air.⁶

Wet kilns typically represent an older cement technology with smaller capacity kilns. In the United States wet cement kiln capacities range from 77,000 to 1,179,000 metric tons/year with an average of 307,000 metric tons/year.⁵

2.1.3.2 Dry Process Kilns

The dry process utilizes a dry kiln feed rather than a slurry. Early dry process kilns were short, and the substantial quantities of waste heat in the exit gases from such kilns were frequently used in boilers to generate electric power which often satisfied all electrical needs of the plant. In one modification, the kiln has been lengthened to nearly the length of wet-process kilns and chains were added. The chains serve almost exclusively a heat exchange function. Refractory heat-recuperative devices, such as crosses, lifters, and trefoils, have also been installed. So equipped, the long dry kiln is capable of better energy efficiency than wet kilns. Other than the need for evaporation of water, its operation is similar to that of a wet kiln. To improve the energy efficiency of the dry process, variations such as suspension preheaters and precalciners have been introduced as discussed in the next sections.⁶

Long dry process kilns are generally of a smaller capacity compared to long wet kilns. In the United States dry cement kiln capacities range from 50,000 to 590,000 metric tons/year with an average capacity of 265,000 metric tons/year.⁵

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2.1.3.3 Dry Kilns With Preheaters

In dry kilns with suspension preheaters, the pulverized feed passes by gravity through a series of cyclones and riser ducts in a vertical arrangement, where it is separated and preheated several times, typically in a four-stage cyclone system. The partially (30 to 50 percent) calcined feed exits the preheater tower into the kiln at about 800 to 900°C (1,500 to 1,650 °F). The kiln length required for completion of the cement formation is considerably shorter than that of conventional kilns, and heat exchange is very good. Suspension preheater kilns are very energy efficient compared to wet or long dry kilns. The intimate mixing of the hot gases and feed in the preheater riser ducts promotes condensation of alkalis and sulfur on the feed, which sometimes results in objectionable high alkali and sulfur contents in the clinker, or occasions buildups in the preheater tower. . To alleviate this problem, some of the kiln exit gases can bypass the preheater through a slip stream, or fewer cyclone stages can be used in the preheater with some sacrifice of efficiency.

Suspension preheater kilns represent a newer cement technology compared to the long dry kilns. They are also somewhat larger in production capacity than the conventional long rotary kilns. In the United States the preheater type kiln capacities range from 223,000 to 1,237,000 metric tons/year with an average capacity of 406,000 metric tons/year.

2.1.3.4 Dry Kilns With Precalciners

The success of preheater kiln systems, led to precalciner kiln systems. These units utilize a second burner to carry out calcination in a separate vessel attached to the preheater. The calciner utilizes preheated combustion air drawn from the clinker cooler or kiln exit gases and is equipped with a burner that typically burns about 60 percent of the total kiln fuel. Most often coal is used as a fuel in a calciner furnace; however, almost any fuel may be used including chipped tires. The raw material is calcined almost 95 percent, and the gases continue their upward movement through successive cyclone/riser duct preheater stages in the same manner as in an ordinary preheater. The precalciner system permits the use of smaller dimension kilns, since only actual clinkering is carried out in the rotary kiln. Energy efficiency is often even better than that of a preheater kiln, and the energy penalty for bypass of kiln exit gases is reduced since only about 40 percent of the fuel is being burned in the kiln. The burning

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process and the clinker cooling operations for the modern dry-process kilns are the same as for long wet kilns.

The precalciner technology is the most modern cement manufacturing technology and almost all of the newer cement plants are based on these designs. Precalciner kilns are also much larger in capacity than the conventional rotary kilns. The precalciner type kilns in the United States range from 449,000 to 1,580,000 metric tons/year with an average of 869,000 metric tons/year. Because of the new large precalciner plants replacing older and smaller plants, the overall average kiln capacity has been steadily increasing in the United States. It has increased from an average of 239,000 metric tons/year in 1980 to an average capacity of 393,000 metric tons/year in 1989.

3.0 NO_x FORMATION IN CEMENT KILNS

In cement manufacturing, conditions favorable for formation of nitrogen oxides (NO_x) are reached routinely because of the high process temperatures involved. Essentially, all of the NO_x emissions associated with cement manufacturing are generated in the cement kiln systems. Although, there are other heating operations in a cement plant, such as drying of raw feed or coal, often the heat from the kiln exhaust gases is used for these operations making their contribution to NO_x emissions negligible. In cement kilns, NO_x emissions are formed during fuel combustion by two primary mechanisms:

- Oxidation of the molecular nitrogen present in the combustion air which is termed thermal NO_x formation, and
- Oxidation of the nitrogen compounds present in the fuel which is termed fuel NO_x formation.

Sometimes the raw material feed to the kiln may also contain nitrogen compounds which may lead to feed NO_x formation similar to fuel NO_x formation. Because of the high temperatures involved in the burning or clinker formation step, the thermal NO_x formation provides the dominant mechanism for NO_x formation in cement manufacturing. The term NO_x includes both NO and NO₂ species, although NO₂ normally accounts for less than 10 percent of the NO_x

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emissions from a cement kiln exhaust stack.¹ The concentration and emission of NO_x are, however, typically expressed in equivalent NO₂ form.

3.1 Thermal NO_x Formation

Fuel combustion in the kiln burning zone is the primary source of thermal NO_x, formation in cement kilns due to temperatures well above 1,400°C (2,550°F). In contrast, the fuel combustion temperature in a precalciner or in a kiln riser duct is well below 1,200°C (2,200°F), suppressing thermal NO_x formation.⁴ Mainly fuel and feed NO_x may be formed in the secondary firing zone of preheater and precalciner kiln systems. Along with the combustion temperature, the gas-phase residence time and the available oxygen concentration in the high temperature kiln burning zone are important parameters. Longer residence times at the high temperatures will allow more NO_x to be formed. Greater amounts of oxygen in the combustion zone will, of course, lead to greater amounts of NO_x formation. Strategies to reduce NO_x emissions need to be based upon reducing formation of NO_x which may be achieved by reducing combustion temperature, oxygen concentration in the high temperature combustion zone, and the gas residence time at high temperatures.

3.2 Fuel NO_x Formation

Fuel NO_x is formed by the conversion of nitrogen present in the fuel used. A recent survey of the cement industry by Portland Cement Association (PCA) indicates that almost 82 percent of the energy requirement of the cement industry is provided by coal.⁵ Natural gas contributed about 3 percent of the energy demand, oil about 1 percent, and other fuels such as waste solvents provided about 14 percent of the energy. Both oil and natural gas have relatively low fuelbound nitrogen content, whereas coal may contain 1 to 3 percent of nitrogen by weight depending upon the source of coal. Waste-derived fuels (WDF) such as scrap tires, used motor oils, surplus printing inks, dry-cleaning solvents, paint thinners, sludge from the petroleum industry, agricultural wastes such as almond shells, and even municipal biosolids (dewatered sewage sludge) are finding an increasing application in the cement kilns.⁶ The nitrogen content in these fuels may be significant depending on the chemicals included in the waste mix being burned.

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3.3 Feed NO_x Formation

Similar to coal, the raw materials used in cement production may contain a significant amount of nitrogen. In most cases, limestone is the major raw material, with the remainder of the raw mix being composed of clays, shales, sandstones, sands, and iron ores. Fly ash from coal-fired power plants is used as a raw material and may contain substantial amounts of nitrogen compounds, since it is usually derived from coal. Most of these raw material components are sedimentary minerals and they may contain small amounts of chemically bound nitrogen, presumably of organic origin. Various kiln feeds contain appreciable amounts of nitrogen, ranging from about 20 ppm up to as much as 1,000 ppm (as N).⁸ The higher values (>100 ppm) are generally associated with minerals displaying significant kerogen contents. Since 100 ppm N in a kiln feed is equivalent to about 1 lb NO_x per ton of clinker (if it all converted), NO_x emissions from the kiln feed may represent a major source of NO_x from cement kilns. Nevertheless, it is probably less important than thermal NO_x and fuel NO_x in most cases.

3.4 Energy Efficiency of the Cement-Making Process

Since NO_x formation is directly related to fuel combustion, any reduction in the amount of fuel burned per unit amount of clinker produced should reduce NO_x emissions per unit clinker. Attempts to improve energy efficiency of the process by avoiding excessive clinker burning and utilizing waste heat effectively for preheating combustion air, coal, and raw mix is likely to reduce NO_x emissions. Improving heat transfer between hot gases and solid materials, e.g., by chain systems, will improve energy efficiency. The newer preheater and precalciner kiln designs provide very efficient preheating and precalcining of the raw mix with intimate gassolids contact in cyclone towers. New cement kiln installations or renovations of older kilns thus predominantly involve precalciner designs for their energy efficiency. The inherent energy efficiency of these kiln designs is likely to produce lower NO_x emissions per unit amount of clinker as compared to the wet or long dry kilns.

4.0 OVERVIEW OF POTENTIALLY APPLICABLE CONTROL TECHNIQUES AND TECHNOLOGIES

The initial part of this project involved a literature review and research potential NO_x control technologies that may be applicable to the differing kiln and processes in Ellis County.

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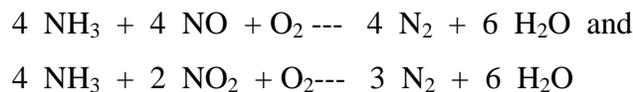
Combustion and process optimization techniques are the primary methods of NO_x control currently in use in the United States. These techniques are fairly well understood and documented for application to US cement plants and are currently in use to some extent at all of the Ellis County cement plants. The greatest area of uncertainty at the onset of this project was related to three technologies that have potential for application to Ellis County kilns, but have had limited or no history of application in the US cement industry. For this reason, the research in Phase I was largely focused on assembling information for assessment of the these technologies. These technologies are:

Selective Catalytic Reduction (SCR),
Selective Non-catalytic Reduction (SNCR) and
Low Temperature Oxidation.

4.1 Selective Catalytic Reduction (SCR)

SCR is a process that uses ammonia in the presence of a catalyst to selectively reduce NO_x emissions from exhaust gases. This technology is widely used for NO_x abatement in other industries, such as coal-fired power stations and waste incinerators. The SCR process has been used extensively throughout the world to achieve a 90 percent reduction in NO_x emissions from fossil fuel fired boilers. In the United States SCR technology has successfully been used for gas turbines, internal combustion engines, and utility boilers.

In SCR, anhydrous ammonia, usually diluted with air or steam, or urea, is injected through a grid system into hot flue gases that are then passed through a catalyst bed to carry out NO_x reduction reactions. The two principal reactions are:



The first reaction represents the predominant reaction since 90 to 95 percent of NO_x emissions in the flue gas are in the form of NO. A number of materials have been used for catalysts. Titanium dioxide (TiO₂) and vanadium pentoxide (V₂O₅) mixtures are most commonly used as catalysts due to their resistance to SO_x poisoning. Zeolite-based catalyst materials have also been developed capable of operating at higher temperatures than

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conventional metal-based catalysts. The catalyst is typically supported on ceramic materials, e.g., alumina in a honeycomb monolithic form. The active ingredients of the base metal (titania-vanadia) and zeolite catalysts often make up the bulk of the substrate material. The catalyst shape and reactor design vary depending upon the manufacturer.

The optimum temperature for the catalytic reactions depends upon the specific catalyst used and is usually in the range between 300 to 450°C (570 to 840°F). Without pre-heater and/or pre-calciner systems this temperature range may be higher than typical cement kiln flue gas temperatures, especially in plants using heat recovery systems or baghouses for particulate collection. It may be possible to reheat the exhaust using heat recovery systems; this can have an impact on cost of control. This is of particular relevance for potential application of SCR at wet kilns.

Ammonia is typically injected to produce a NH_3 : NO_x molar ratio of 1.05-1.1:1 to achieve NO_x conversion of 80 to 90 percent with an ammonia "slip" of about 10 ppm of unreacted ammonia in the gases leaving the reactor. The NO_x destruction efficiency depends upon the temperature, NH_3 : NO_x molar ratio, and the flue gas residence time (or the space velocity) used in the catalyst bed. The SCR reactor system can be designed for a desired NO_x reduction using appropriate reagent ratio, catalyst bed volume, and operating conditions. In general, the catalysts may be fouled or deactivated by the particulates present in the flue gas. In the case of cement plants, the presence of alkalis and lime, as well as sulfur dioxide, in the exhaust gases is also of concern. Recent developments, however, have led to more sulfur tolerant SCR catalysts. In addition, soot blowers may be used to prevent dust accumulation on SCR catalysts.

In the cement industry, basically two SCR systems are being considered: low dust exhaust gas and high dust exhaust gas treatment. Low dust exhaust gas systems require reheating of the exhaust gases after de-dusting, resulting in additional cost. High dust systems are considered preferable for technical and economical reasons.

SCR may only be available as an applicable technology for dry kilns with preheat, as is described later herein. There are currently no installations of SCR units in any United States cement plants, however, a full-scale unit has been operating in Germany for over three years with SCR. As set forth in greater detail below, selective catalytic reduction has been installed and successfully operated at this one cement plant in Germany and SCR is now commonly used

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to control NO_x emissions from coal-fired power plants. This technology achieves NO_x emission reductions in the range of 80 to 90 percent or higher.

The experience of the German cement plant may be sufficient to demonstrate that the concerns regarding catalyst poisoning and fouling have been addressed with the development of a special catalyst specifically designed for the cement industry. In a typical design of a dry kiln with preheat, the SCR system could be located downstream of the pre-heater cyclones, prior to the roller mill. The process exhaust temperature at the outlet of the pre-heater cyclones would be approximately 320°C (608°F), which is ideal for a SCR system.

Potential Advantages of SCR. The potential advantages of SCR are as follows:

- **SCR can potentially provide a high degree of NO_x removal.** Presently, guarantee levels on SCR systems being provided in the power industry are in the 90% to 94% range. Even on difficult applications such as coal-fired power plants, which have very dusty flue gases, NO_x control efficiencies of 90%+ are being guaranteed by vendors and being achieved.

SCR is an add-on or end-of-pipe control technology. For dry kilns with preheat, the SCR system would be located downstream of the cement kiln and pre-heater furnace and cyclones and hence would minimize interference with the cement manufacturing process.

- **SCR can use urea (CO(NH₂)₂) rather than ammonia as the reducing agent.** To use urea, a conversion system is required. In a typical conversion system, urea is mixed with water and then heated. The process converts the urea to gaseous ammonia, which is then injected into the flue gas. This system is commonly used where there are safety concerns arising from the transport and storage of ammonia.

SCR Experience in Cement Kilns. The Solnhofer Portland-Zementwerke AG cement plant in southern Germany located between Munich and Nuremberg has been in successful operation with SCR since 2001. Lurgi, the engineering company who supplied the SCR system, addressed the problem of catalyst deactivation discussed above (caused by the high calcium content of cement plant ash) by developing a catalyst specifically for the cement industry. The catalyst can use ammonia or urea as a reagent. If urea is used, a conversion unit is used to convert the urea to ammonia prior to injection in the SCR system. These units, which are quite common, avoid the need to store ammonia on-site for use as a reagent.

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As of the beginning of 2005, there was no evidence of catalyst poisoning or deactivation. The problem of fouling from the heavy dust loading has proven to be manageable with regular maintenance (i.e., cleaning of the catalyst with a vacuum cleaning system).

The SCR system is achieving NO_x emission reductions far in excess of those achievable using SNCR. Prior to installation of SCR, the Solnhofer plant, with SNCR, was achieving NO_x emissions of 700 to 800 mg/ Nm³ (equivalent to 2.8 to 3.2 lb/ton of clinker). With SCR, the plant has been achieving approximately 200 mg of NO_x/Nm³ (0.8 lb/ton of clinker). Based on an uncontrolled NO_x emission rate of 1050 mg/Nm³ (4.2 lb/ton of clinker), the emission rate corresponds to a NO_x control efficiency of 80%. It has been indicated, however, that the SCR system was capable of achieving a lower NO_x emission rate (they are not using all of their beds), but that they were operating at 200mg/Nm³ since German law only required a NO_x emission limit of 500mg/Nm³.

Areas of Concern. A number of areas of concern have been raised with respect to SCR utilization for US cement kilns. Cost of control has traditionally been a concern with SCR including high installation costs due to difficulty of installation and the need to optimize gas steam conditions. In addition to cost, several key issues are being more fully investigated under Phase II of this study. These issues include:

Catalyst Deactivation.

Poisoning of Catalyst.

SCR Catalyst Plugging and Fouling.

Catalyst Composition, NO_x Reduction Efficiency and SO₂ Oxidation.

Gas Temperature Ranges, Distribution and Fluctuations.

NO_x Inlet Concentration Variability and NH₃ Slip.

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Undesirable Byproduct Formation and Adverse Impact of Byproducts on
Downstream Equipment.

Process Start-up, Shutdown and Malfunction Events.

Gas Flow Distribution.

Based on what has been studied to date, it appears that SCR should be considered as a potentially available technology for dry kilns with pre-heaters and/or pre-calciners, especially with the high control efficiencies of 90% or greater control of NO_x achieved with coal-fired utility boilers. With respect to wet kilns, the exit gas temperatures typically would necessitate raising of the kiln gas temperature prior to entering an SCR system on the order of 200 F. This would have a significant cost that may preclude its utilization on economic grounds. In both cases, however, SCR utilization for retrofits must be studied on a case-by-case basis considering the site specific characteristics of each specific kiln.

4.2. Selective Noncatalytic Reduction (SNCR)

This control technique relies on the reduction of NO_x in exhaust gases by ammonia or urea, without using any catalyst, using the same reactions as in the case of the SCR process. This approach avoids the problems related to catalyst fouling, as in SCR technology, but requires injection of the reagents in the kiln at much higher temperatures, in the range between 870 to 1,090°C (1,600 to 2,000°F). At these temperatures urea decomposes to produce ammonia which is responsible for NO_x reduction. In principle, any of a number of nitrogen compounds may be used, e.g., cyanuric acid, pyridine, and ammonium acetate. However, for reasons of cost, safety, simplicity, and by-product formation, ammonia and urea have been used in most of the SNCR applications.

Because no catalyst is used to increase the reaction rate, the temperature window is critical for conducting this reaction. At higher temperatures, the rate of a competing reaction for the direct oxidation of ammonia which actually forms NO_x becomes significant. At lower temperatures, the rates of NO_x reduction reactions become too slow resulting in too much

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unreacted ammonia slip. The effective temperature window range can be lowered to about 700°C (1,300°F) by the addition of hydrogen along with the reducing agent.⁵¹ Nalco Fuel Tech, the producer of the SNCR technology NO_xOUT[®], has also introduced NO_xOUT[®] PLUS which is said to broaden the operating temperature window and to reduce ammonia slip and CO and NO₂ formation.

In a conventional long kiln the appropriate temperature window is in the middle of a kiln. Because of the rotating nature of a long kiln, continuous injection of ammonia- or urea-based reagents has presently not been considered possible. The technology developed for mid-kiln firing of containerized solid fuels allows injection of a certain amount of material once during the kiln revolution. Injection of solid ammonium or urea salts in this manner has not been used because of the rapid decomposition of such salts. Therefore, SNCR technology has not been applicable for long dry or wet kilns. However, Fuel Tech Inc. is currently evaluating dry dust or pellet injection methods for direct injection into kilns.⁵² The possibility of injection of ammonia or solid urea inside automobile tires that are injected into the kiln has still to be evaluated. With the known ability of mixing air fans to induce turbulence in the system, mixing combustion gases containing NO_x with ammonia, either generated from urea or from ammonium salts, may still offer possibilities. Chain gas temperatures generally are in the range of 1500-1600°F (815-871°C). The mid-kiln injection systems are usually down kiln somewhat from this point, which means that they will be higher than this in gas temperature. There is a good possibility that they will be in the right range, if mixing is induced. This option needs to be explored.

In preheater/precalciner type cement kilns, the temperatures at the cooler end of the rotating kiln, in the riser duct, and in the lower section of the cyclone preheater tower are likely to be in the temperature window appropriate for SNCR. Such kilns are therefore good candidates for application of SNCR technology.

SNCR is presently being used in 18 cement kilns in Europe. Fifteen kilns are in Germany, two are in Sweden, and one is in Switzerland. These kilns are either suspension preheater kilns or precalciner kilns. The most common reagent used is 25% ammonia water.¹ NO_x reduction rates vary from 10 to 50 percent with NH₃/NO₂ molar ratio of 0.5 to 0.9, NO_x emissions at these reductions are 2.4 to 3.8 lb/ton of clinker. Two dry process cyclone preheater/precalciner kilns in Sweden achieve 80 to 85 percent reduction (1.0 lb/ton of clinker) at a NH₃/NO₂ molar ratio of 1.0 to 1.1.¹ One reason for the remarkable results in these kilns is the

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use of multiple injection points (12 in all) that ensure long enough retention time for the reaction to take place. The optimum temperature is said to be 900-1,100°C (1,650-2,010°F). These results suggest that some SNCR tests may have been carried out under lower-than-optimum conditions.

F. L. Smidth and Company tested SNCR on a preheater/precalciner kiln³ Ammonia was injected into the lower cyclone of the preheater tower where temperatures are favorable for the reduction reactions to occur. NO_x emissions reductions during this experiment averaged 40 percent, but NO_x reductions of over 90 percent were obtained when the ammonia injection rate was 10 to 20 percent in excess of stoichiometric.

In North America, SNCR has been tested on at least seven kilns (this is increasing rapidly at the present time and needs to be updated). In some of the earlier work, NOxOUT[®] technology was tested under ten different operating conditions at a preheater/precalciner kiln in Seattle, Washington during October 1993. NO_x emissions were effectively reduced from 3.5 to 6.0 lb NO_x/ton of clinker to less than 1 lb NO_x/ton of clinker.^{57, 58} Another test of NOxOUT[®] technology was conducted during October 1998 in Davenport, Iowa. This test found NO_x reductions of 10 to 20 percent from a baseline of approximately 350 pounds NO_x/hour, although higher levels of reduction are thought to be achievable when the baseline is higher.⁵⁹ An evaluation of NOxOUT[®] technology was conducted in 1994 for a long dry kiln in Southern California, but the study concluded that application of the NOxOUT[®] technology at the subject kiln was technically infeasible.⁶⁰ The details of this testing are not known. One of the Ellis County cement plants has been evaluating SNCR. *INSERT TEXT HERE* Present evidence appears to favor the use of ammonia over urea. A modern Florida precalciner plant recently obtained very good, almost stoichiometric reduction of already low NO_x levels using 19% aqueous ammonia. There are at least two variant technologies that of the basic SNCR techniques described above that will be evaluated in this project, these include:

Biosolids Injection (BSI).

NOxOUT[®].

4.3 Low Temperature Oxidation Technologies

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4.3.1 Introduction

Oxidation technologies for flue gas NO_x control developed in recent years have become commercially successful and economically viable as an alternative to ammonia and urea based technologies using reduction chemistry to remove NO_x. Older commercial technologies such as Selective Catalytic Reduction (SCR) and Selective Non-Catalytic Reduction (SNCR), which reduce NO_x to nitrogen using ammonia or urea as an active chemical, are sometimes limited in their use for high particulate and sulfur containing NO_x streams such as from coal-fired combustors, or are unable to achieve sufficient NO_x removal to meet new NO_x regulation levels. In contrast, oxidation technologies convert lower nitrogen oxides such as nitric oxide (NO) and nitrogen dioxide (NO₂) to higher nitrogen oxides such as nitrogen sesquioxide (N₂O₃) and nitrogen pentoxide (N₂O₅). These higher nitrogen oxides are highly water soluble and are efficiently scrubbed out with water as nitric and nitrous acids or with caustic solution as nitrite or nitrate salts. NO_x removal in excess of 90% has been achieved using oxidation technology on NO_x sources with high sulfur content, acid gases, high particulates and processes with highly variable load conditions. Generally these technologies work at relatively low temperatures (100-950°F) and some of these technologies are likely most cost effective for use on plants with existing scrubbers. These technologies are used for NO_x control for a variety of NO_x emission sources but are not known to have been used to date on a cement kiln.

4.3.2 LoTox

General description. The LoTOx System uses ozone as the oxidizing agent, which selectively oxidizes relatively insoluble NO_x to higher oxides of nitrogen. These higher oxides are readily soluble and easily removed in a wet scrubber. The patented system, which can be installed as a stand-alone or retrofit design, provides sufficient residence time under maximum load conditions to allow complete reaction of the ozone. The system's small and adaptable footprint, as well as its ability to produce ozone on demand, provides customers with optimal control of the installation to maximize NO_x control and minimize capital and operating costs. The LoTOxTM System is very selective for NO_x removal, oxidizing only the NO_x and therefore efficiently using the treatment chemical, ozone, without causing any significant SO_x oxidation and without affecting the performance of the downstream SO_x scrubber.

Figure 4-5 is an overall process diagram of the LoTOxTM System.

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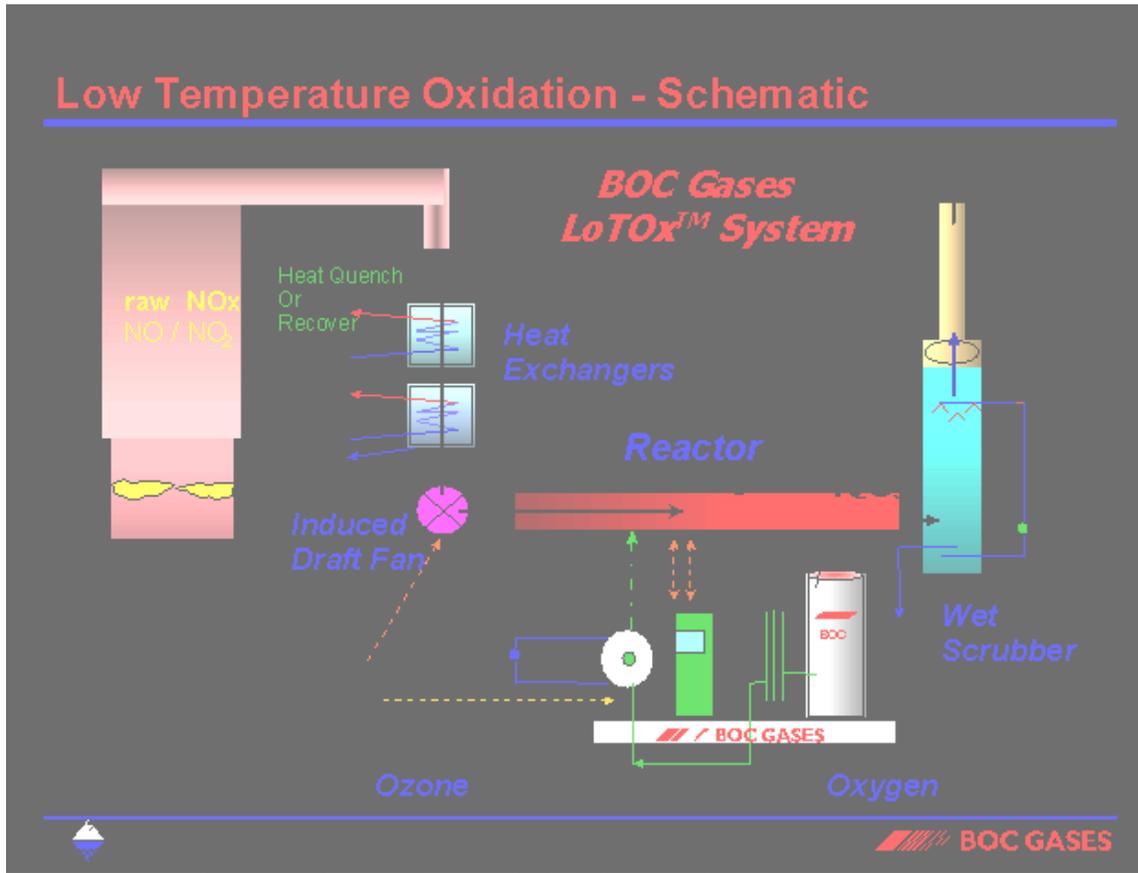


Figure 4-5. Overall Process Diagram of the LoTOx™ System.

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Figure 4-6 provides another example of a LoTox system where the ozone injection is included in the scrubber.

EDV[®] Wet Scrubbing System With LoTOx[™] Injection

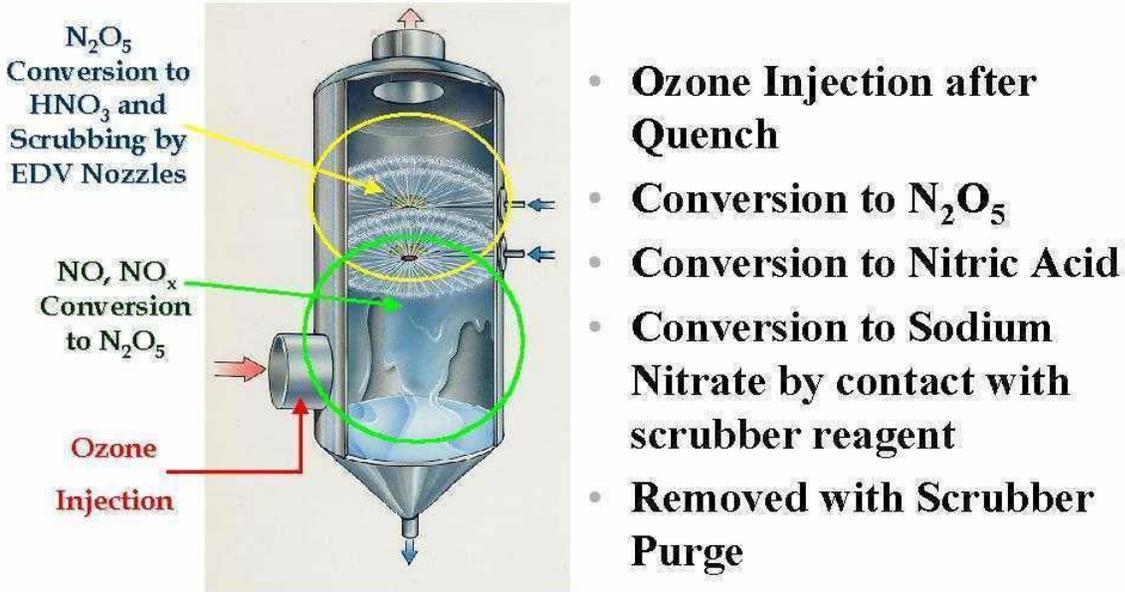


Figure 4-6. EDV Wet Scrubbing System with LoTOx[™] Injection.

Potential applicability. The applicability of the LoTox system may be limited by the large gas volumes, high moisture levels and large quantities of dilution air needed to reach the operating temperature range in some systems. (Further investigation is needed.)

Advantages

Table 4-9. Comparison Of Common Post Combustion NO_x Abatement Technologies For NO_x Emissions Control

	SNCR	SCR	LoTOx [™]
Mode of Treatment	Reduction	Reduction	Oxidation
Active Chemical	NH ₃ / Urea	NH ₃ / Urea	Ozone
Gas Temperature Required °F	1650 - 2000	500 - 900	150 – 250
Pressure Required, psig	0+	0++	0+

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Placement in Exhaust System	Near Combustion Chamber	Between Air Pre-Heater and Economizer (boiler)	Tail End
Catalyst Bed	No	Yes	No
Gas Phase Reaction Duct	Yes	No	Yes
Scrubber	Optional	Optional	Yes
NOx Reduction Achieved	40 - 70 Percent	60 - 95 Percent	90 - 98 Percent
Slip of Active Agent	NH ₃ – Yes	NH ₃ - Yes	Ozone - No
CO Emissions After Treatment	May Increase	May Increase	No Effect
SOx Emissions After Treatment	Little Effect – Maybe H ₂ S	Little Effect - Maybe H ₂ S	No Effect or Significantly Reduced
Gas Temperature Outside Operating Range - Overshoot	More NOx Emissions Through NH ₃ Oxidation	More NOx Emissions Through NH ₃ Oxidation, Ammonia Slip	Increased O ₃ Consumption for the Time Period of Overshoot
Gas Temperature Outside Operating Range - Undershoot	More NOx Emissions Through Reduced Reduction by NH ₃	More NOx Emissions Through Reduced Reduction by NH ₃ , Ammonia Slip	No Effect

Advantages. One of the key advantages of the oxidation technologies is their lower operation temperature ranges. This allows installation in locations that are not optimal for higher temperature SCR and SNCR applications. In addition, the oxidation technologies can also control additional pollutants. Although the chemical composition of the stream can influence the specifics of control, it is likely that oxidation technologies will also oxidize and remove other gaseous pollutants such as organic hydrocarbons and oxides of sulfur from the gas steams.

Disadvantages. It is likely that for most application the LoTox option would be an add-on to existing APCD systems. Because of high electrical power usage costs are reported to be relatively high. These systems oxidize oxides of nitrogen so that they can be hydrated and scrubbed from the gas steam. This requires installation of expensive wet scrubbers for plants that are not currently equipped with scrubbers.

In addition to the LoTox process described above, there are other oxidation technologies that will be evaluated in this report. These include:

Hydrogen Peroxide

OXONE (Dupont – potassium monopersulfate)

Sodium Acetate Process

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REFERENCES

This report is a draft interim public version of the internal mid-project draft report developed for TCEQ. No attempt was made to develop a set of references for this draft interim public version of the report. However, the numbered references are believed to be consistent with those of the EPA Report titled “NO_x Control Technologies for the Cement Industry”, U.S. Environmental Protection Agency Office of Air Quality Planning and Standards, September 19, 2000 which was used as the primary source of information for Section 2 and 3 and parts of Section 4 of this report.

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ATTACHMENT 1 – September 19, 2000 EPA Report - *NO_x Control Technologies for the Cement Industry*

Table 1-1. ERG’s Summary of the September 19, 2000 EPA Report *NO_x Control Technologies for the Cement Industry*

NO _x Control Strategies		Applicability	NO _x Reduction Efficiency (%)	Estimated Costs	Number of Installations
Process modifications	Excess air and temperature optimization (using CO and O ₂ CEMS and feedback control)	All kiln types	15 ^a	Capital cost = ~\$750,000 ^s	Unknown
	Reduce alkali content of feed where feasible	All kiln types	Not available	Not available	Unknown
	CemStar (addition of steel slag)	All kiln types although higher NO _x reductions expected from wet and long-dry kilns due to greater heat input offsets	9 - 60 ^b	Capital cost = \$200,000 - \$500,000 ^t Annual costs are highly site-specific (e.g., slag injection rate, potential cost savings due to increased production)	11
	Fuel switching (natural gas to coal)	All kiln types	70 ^c	Not available	~ 87% of existing kilns use coal
	Improving thermal efficiency	All kiln types	Not available	Not available	Unknown
Combustion modification	Staged air combustion - staged fuel combustion	All kiln types (most effective with indirect fired kilns)	Not available	Not available	Unknown
	Staged air combustion - flue gas recirculation	May not be suitable for cement kilns. Incorporation of FGR in a cement kiln also results in somewhat increased power consumption and reduced kiln output.	Not available	Not available	Unknown

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Table 1-1. ERG’s Summary of the September 19, 2000 EPA Report *NO_x Control Technologies for the Cement Industry*

NO _x Control Strategies		Applicability	NO _x Reduction Efficiency (%)	Estimated Costs	Number of Installations
	Staged air combustion - flue gas recirculation w/low-NO _x burner		15 - 38 ^d	Not available	Unknown
	Staged air combustion - Low-NO _x burner	All kiln types (burners that use 5-7% excess air are only for indirect-fired kilns)	44 ^e 15 - 33 ^f 23 - 47 ^g 14 ^h	Capital cost = \$511,000 - \$966,000 ^u Annual cost = \$136,000 - \$204,000 ^u	~ 22 existing kilns use low-NO _x burners
	Staged fuel combustion - Tire-derived fuel (TDF)	All kiln types	30 - 40 ⁱ		Unknown. Approximately 53,300,000 tires are burned in cement kilns annually http://www.epa.gov/epaoswer/non-hw/muncpl/tires/tdf.htm#cement
	Staged fuel combustion - Low-NO _x precalciner	Not applicable to preheater kilns that fire fuel in the riser	≤46 ^j	Not available	Unknown
	Staged fuel combustion - Mid-kiln firing	Wet and long-dry kilns	38 ^k 11 - 55 ^l 28 - 59 ^m	Capital cost = ~ \$3,000,000 ^v Annual cost = (\$370,000) - \$189,000 ^v	21 long kilns (US), 40 kilns (worldwide)
Post-formation removal	Selective catalytic reduction (SCR)	No cement kilns had SCR at the time of the EPA Study prior to 2000.		Not available in 2000.	No installations were reported on cement kilns in EPA 2000 report.
	Selective non-catalytic reduction (SNCR)	Best application is for preheater/precalciner kilns. Generally not applicable to long-dry or wet kilns	71 -98 ⁿ 10-20 ^o	Not available	2 kilns (US), 18 in Europe

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Table 1-1. ERG’s Summary of the September 19, 2000 EPA Report *NO_x Control Technologies for the Cement Industry*

NO _x Control Strategies		Applicability	NO _x Reduction Efficiency (%)	Estimated Costs	Number of Installations
	Biosolids (SNCR)	Preheater/precalciner kilns	Not available	Capital cost = \$240,000 ^w Annual cost = (\$322,000) ^w	Unknown
	NO _x OUT (SNCR)	Preheater/precalciner kilns	> 50 - 90 ^p 10 - 20 ^q 46 - 53 ^r	Capital cost = ~\$1,200,000 ^x Annual cost = \$560,000 - \$2,000,000 ^x	None
	Low Temperature Oxidation (LoTox, Hydrogen Peroxide, OXONE (Dupont) & Sodium Acetate)	Oxidation technologies were not discussed or considered in the EPA 2000 report.	Not considered	Not considered	Oxidation technologies were not discussed or considered in the EPA 2000 report.

^aResults based on experimental test on one kiln.

^bResults derived from short- and long-term tests on two different kiln types.

^cResults based on emission tests of dry kilns.

^dResults based on a single 1988 study. The percent reduction depends on degree of flue gas recirculation.

^eResults based on emission tests of a preheater/precalciner kiln.

^fResults based on emission tests of five kilns.

^gResults based on emission tests of four kilns.

^hEmission test of one kiln.

ⁱResults based on tests conducted at one California facility.

^jResults based on a test conducted by the Portland Cement Association.

^kResults based on one study.

^lResults based on testing of seven dry kilns.

^mResults based on testing of three wet kilns.

ⁿBased on emission tests of a preheater/precalciner kiln at one facility.

^oResults based on emission test at one facility

^pResults based on emission tests of a single kiln.

^qResults based on a emission tests of a single kiln (operating conditions during most of the test were unstable).

^rResults based on emission test of two kilns.

^sEstimate for a commercially available process control system.

^tCost estimates obtained from Texas Industries (TXI).

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Table 1-1. ERG's Summary of the September 19, 2000 EPA Report *NO_x Control Technologies for the Cement Industry*

NO _x Control Strategies	Applicability	NO _x Reduction Efficiency (%)	Estimated Costs	Number of Installations
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^uCosts derived for retrofitting low-NO_x burners to eight model kilns with clinker production capacities ranging from 25 to 150 tons of clinker per hour.

^vCosts derived for retrofitting low-NO_x burners to four model kilns with clinker production capacities ranging from 25 to 50 tons of clinker per hour. Annual costs reflect the fuel credit associated with using tire-derived fuel.

^wCosts based on a clinker production rate of 215 tons per hour.

^xCosts based on two preheater/precalciner kilns with a clinker production rates of 92 tons per hour and 133 tons per hour.

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