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Title: NOX REMOVAL IN JET-ENGINE TEST CELL EXHAUST:
PROPOSED NON-THERMAL PLASMA SYSTEMS AND
ECONOMIC CONSIDERATIONS

Author(s): Louis A. Rosocha, LANL/P-24
Jen-Shih Chang, Kuniko Urashima, and Seock J. Kim
McMaster University,
Andrzej W. Miziolek, Michael J. Nusca, and Robert G. Daniel
US Army Research Laboratory,
Robert F. Huie and John T. Herron
National Institute of Standards and Technology

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L.A. Rosocha

Los Alamos National Laboratory

J.-S. Chang, K. Urashima, and S.J. Kim

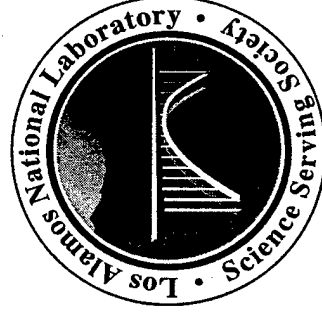
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* This work supported in whole or in part by the U.S. Strategic Environmental Research and Development Program (SERDP).

Thanks to

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Program (SERDP)

for support for this work.

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Outline of talk

- Motivation for and applications of research
- Background of atmospheric-pressure, non-thermal plasma processing for environmental applications (air-pollution control)
 - ♦ Basics of radical formation & decomposition chemistry
 - ♦ Figures-of-merit & costs for decomposition of example pollutants
- Removal of NO_x and VOCs from jet-engine test facilities
 - ♦ Plans for field demonstration of pilot unit
 - ♦ Candidate NTP systems
 - ♦ Economic considerations (candidate systems compared to conventional)
- Future trends
- Summary

Technology-development motivators are regulatory, economic, and social

Regulatory

- New and/or more stringent regulations
Clean Air Act Amendment of 1990, Clean Water Act
Attendant MACT standards

- Enforcement of regulations

Economic

- Impact of regulations
- Costs associated with conventional methods

Social

- Environmental consciousness
- Health risks

The CAAA 1990 contains three titles closely related to NTPs

Title III (Air Toxics)

- Directly concerned with emission of hazardous air pollutants (HAPs)
- Established standards for 189 listed chemicals

Title IV (Acid Rain)

- Reduction of SOx/NOx emissions

Title VI (Stratospheric Ozone)

- Phase out of VOCs

MACT (Max Achievable Control Technology) Standards

- Calls for state-of-art pollution control equipment
- Require changes in equipment, processes, treatment

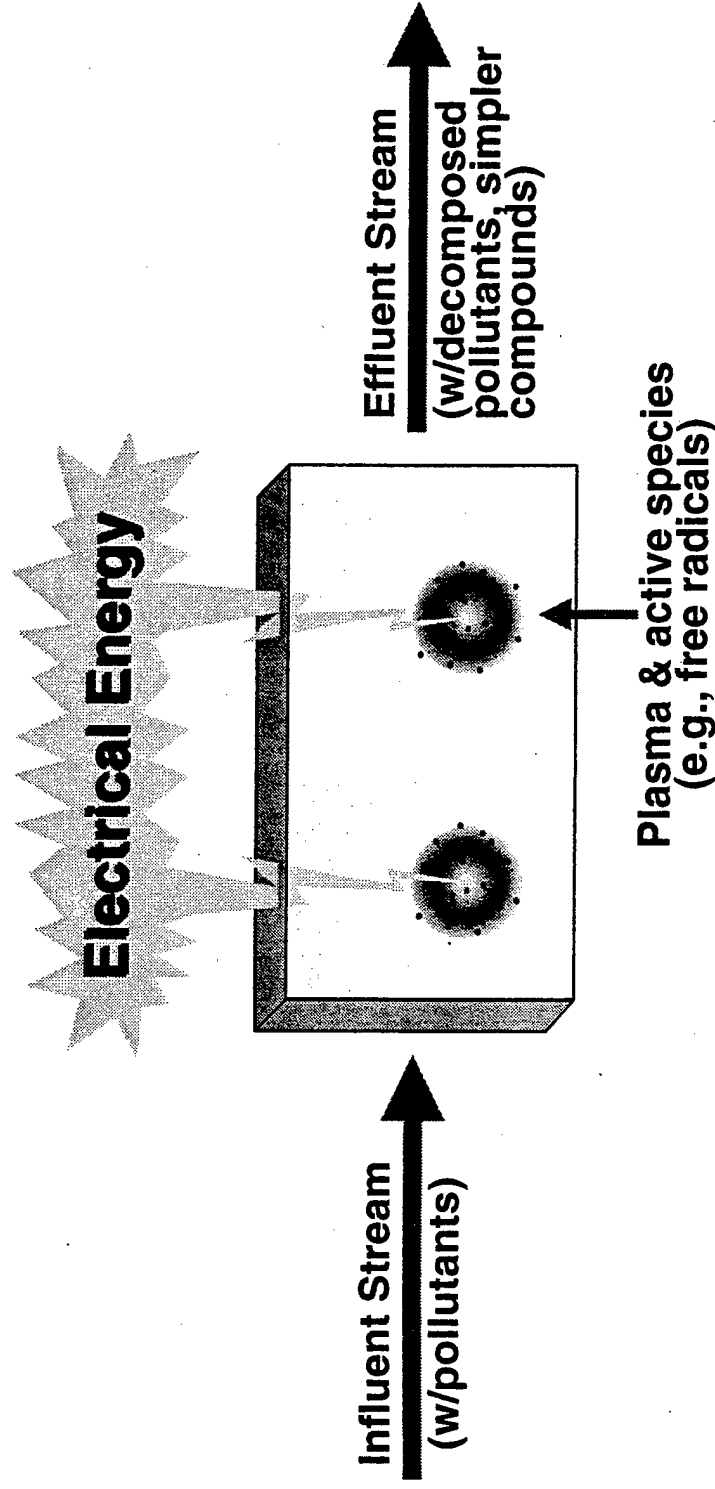
Regulations for engine test facilities are changing under CAAA 1990 10-yr provisions

- Previously, engine test facilities were considered mobile sources because engines were moved in and out of the facilities for testing operations. However, the test facility itself was usually a permanent structure.
- The upcoming new standards will consider the test facilities as stationary sources, which are regulated differently (more stringently) than mobile sources.
- The test-facility emissions (primarily NO_x and volatile organic compounds - VOCs) have dilute concentrations (e.g., < 50 ppm) and very high ($\sim 10^6 - 10^7$ Nm³/h) exhaust-gas flows.
- Consequently, new emissions-control technologies are being explored.

Non-thermal plasmas (NTPs) are a type of advanced oxidation & reduction process which use free radicals to decompose pollutants in the gas phase.

Non-thermal plasmas are a type of AOP making use of “cold combustion” via free-radical reactions. Electrons are energetic (<i>hot</i>), while neutral & ions are at near-ambient temperatures (<i>cold</i>).	Applications • Flue gases & engine emissions: e.g., NO_x & SO_x • VOCs: e.g., hydrocarbons & halocarbons • Odors: H₂S, others
The key idea is to direct electrical energy into favorable chemistry for oxidizing and/or reducing pollutants to more manageable forms (simpler or mineralized terminal products).	Potential Advantages • In-situ generation of chemical reactants • No added fuel (greenhouse gases) • Simultaneous removal of multiple pollutant species • Electronic feedback for optimal process control.

Non-thermal Plasmas Decompose Pollutants Via Active Species Generated in the Process Gas



The radical production efficiency (G-value) depends on the gaseous electronics/plasma chemistry.

$$G = f \left(\frac{k_{rad}}{V_d} \frac{E}{N} \right)$$

- Radical generation is mainly initiated by energetic-electron collisions,
- E/N is the reduced field,
- V_d is the electron drift velocity, which depends on E/N ,
- k_{rad} is the rate constant for radical formation (e.g., a dissociation rate constant, which depends on E/N), and/or other rate constants.

The overall process efficiency can be effectively separated into two terms: the radical production efficiency and the radical utilization efficiency.

$$\eta_{\text{net}} = G_R \eta_u$$

- G_R is the radical-production efficiency (mainly depends on gaseous electronics/plasma chemistry)
- η_u is the radical utilization efficiency (mainly depends on chemical kinetics)

Gas-phase plasma chemical decomposition is driven by electron impact and radical attack.

(1) $e + X \rightarrow \text{products}$

(2) $O, OH, N, \text{etc.} + X \rightarrow \text{products}$

- The first reaction is dominant at large pollutant mole fractions.
- The second dominates at smaller mole fractions.

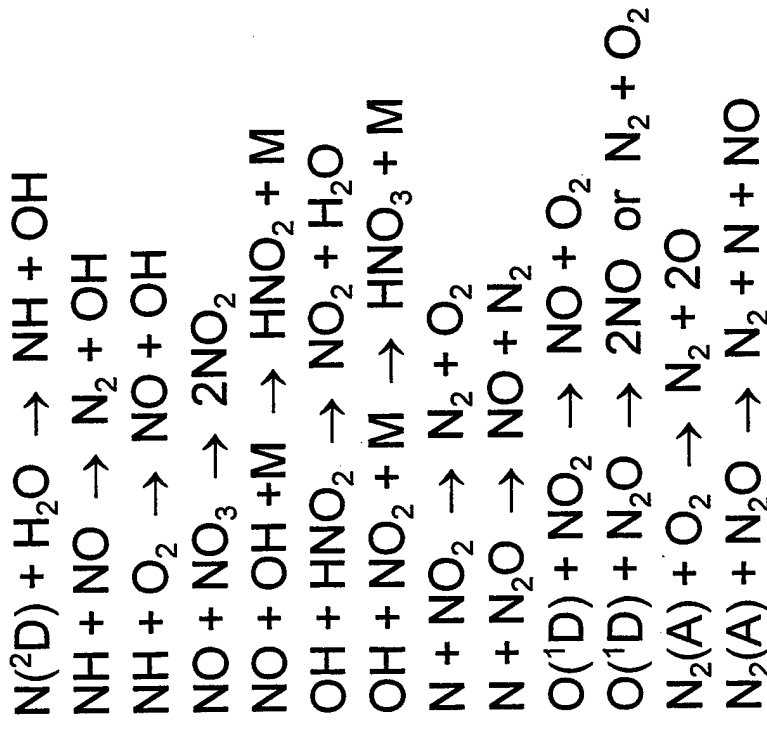
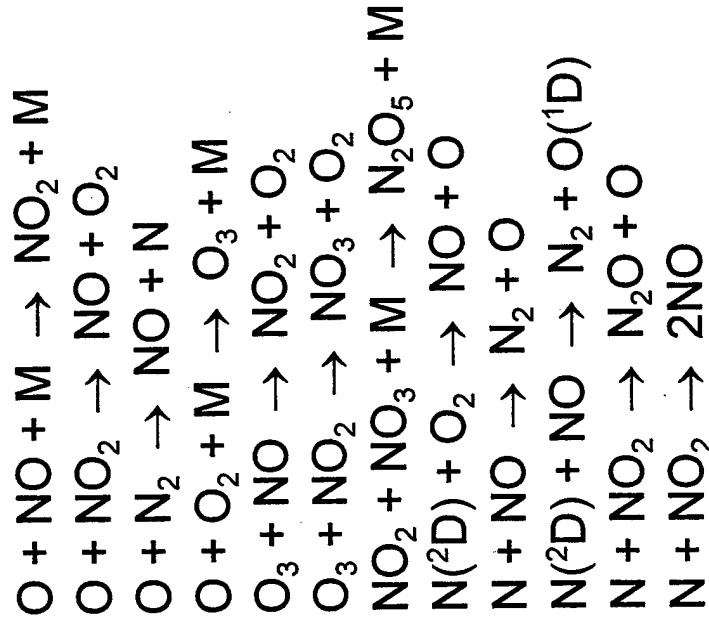
(More energy is directly absorbed by pollutant at high mole fraction, hence electron channel dominates.)

Plasma chemical decomposition of VOCs produces a variety of terminal products.

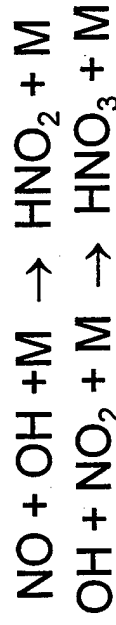
$e, O, OH, N, \text{etc.} + X \rightarrow \text{products}$

- **Manageable products:** $CO_2, CO, Cl_2, HCl, COCl_2$
- **Undesirable products:** Other halocarbons, hazardous byproducts (e.g., DCAC - $CHCl_2COCl$); polymers
- **Secondary treatment:**
 $CO_2 + NaOH \rightarrow NaHCO_3$
 $CO + \text{catalyst} \rightarrow CO_2$
 $Cl_2 + NaHCO_3 \rightarrow NaCl + HCl + \dots$
 $HCl + NaOH \rightarrow NaCl + H_2O$
 $COCl_2 + H_2O \rightarrow 2HCl + CO_2$

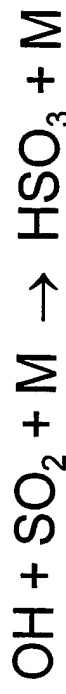
Major de-NO_x Reactions in Moist Gas Mixtures w/o HCs



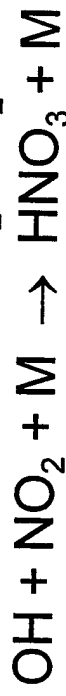
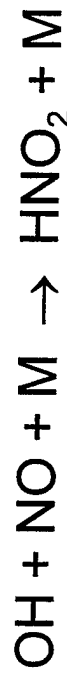
Primary Acid-Formation Pathways:



The presence of SO₂ recycles OH radicals & reduces effective de-NO_x energy cost



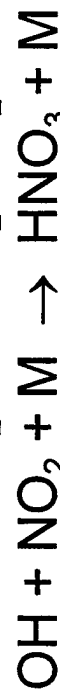
The OH radical then goes on to be used again in de-NO_x



Acid is also formed by the reaction



With ammonia (NH₃) addition, useful particulates (fertilizer) can be formed from NO_x



A generalized removal equation depends on plasma chemistry (radical yields), reaction chemistry, and applied plasma specific energy.

Generalized differential equation:

$$\frac{k[X] + \sum_i k_{S_i} [S_i]}{k[X]} d[X] = -G d\bar{E}$$

Integration with limits $[X]_0 \rightarrow [X]$ and $0 \rightarrow E$ gives:

$$\frac{[X]}{[X]_0} + \frac{\sum_i k_{S_i} [S_i]}{k[X]_0} \ln \frac{[X]}{[X]_0} - 1 = -\frac{G\bar{E}}{[X]_0}$$

For low degree of removal (i.e., $[X]/[X]_0 \sim 1 + \ln \{ [X]/[X]_0 \}$), an analytical solution is obtained:

$$[X] / [X]_0 = \exp (-\bar{E} / \beta)$$

where

$$\beta = \frac{1}{G} \left([X]_0 + \frac{\sum_i k_{S_i} [S_i]}{k} \right)$$

When $k[X] \ll \sum_i k_{S_i} [S_i]$, the β -value and, hence the degree of removal $[X]/[X]_0$ shows no dependence on the initial concentration $[X]_0$.

Example scaling laws for pollutant removal (non-sensitive $[X]_0$ dependence)

$$(1) [X] = [X]_0 \exp(-\bar{E}/\beta),$$

where $[X]_0$ is the initial pollutant concentration, $[X]$ is the resulting concentration, $\bar{E}$ is the applied specific energy (or plasma power divided by gas flow rate, P/Q), and β is the e-fold energy density. Supplying one β to the reactor reduces the concentration by $1/e$, 2β by $1/e^2$, and so on.

A useful figure of merit for the decomposition of pollutants is defined by the energy delivered to the plasma per hazardous molecule removed from the gas stream. At any instant, this can be expressed as the following quantity obtained by solving Equation 1 for $\bar{E}$ and taking the derivative:

$$(2) \gamma_i = -\frac{d\bar{E}}{d[X]} = -\frac{d[-\beta \ln(\frac{[X]}{[X]_0})]}{d[X]} = \frac{\beta}{[X]}.$$

This is the instantaneous energy cost per molecule removed.

Scaling laws for pollutant removal (cont'd)

A more practically-useful parameter is the integral, or average, energy cost γ

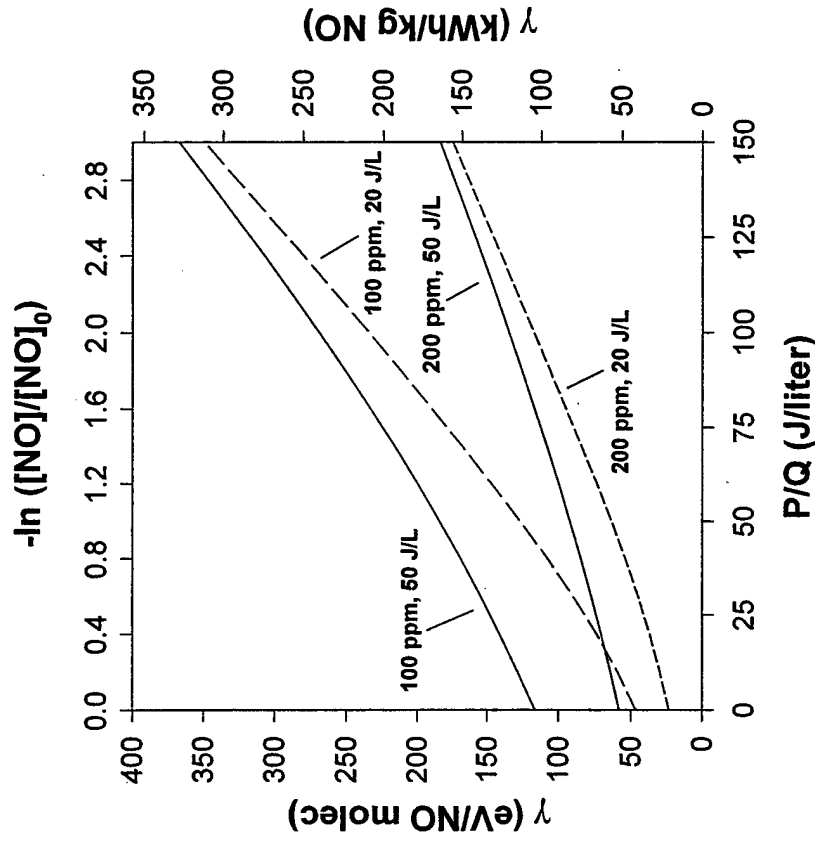
$$(3) \quad \gamma = \frac{\bar{E}}{[X]_0 - [X]} = \frac{-\beta \ln\left(\frac{[X]}{[X]_0}\right)}{[X]_0\left(1 - \frac{[X]}{[X]_0}\right)} \quad (= \beta/[X]_0 \text{ (at optimum; i.e., } [X]/[X]_0 \sim 1)).$$

Here, the energy cost per molecule is expressed in terms of the β -value, the degree of removal, and the initial concentration.

When radical-pollutant attack dominates $\beta \approx [X]_0/G$ and the removal cost is then independent of the initial concentration

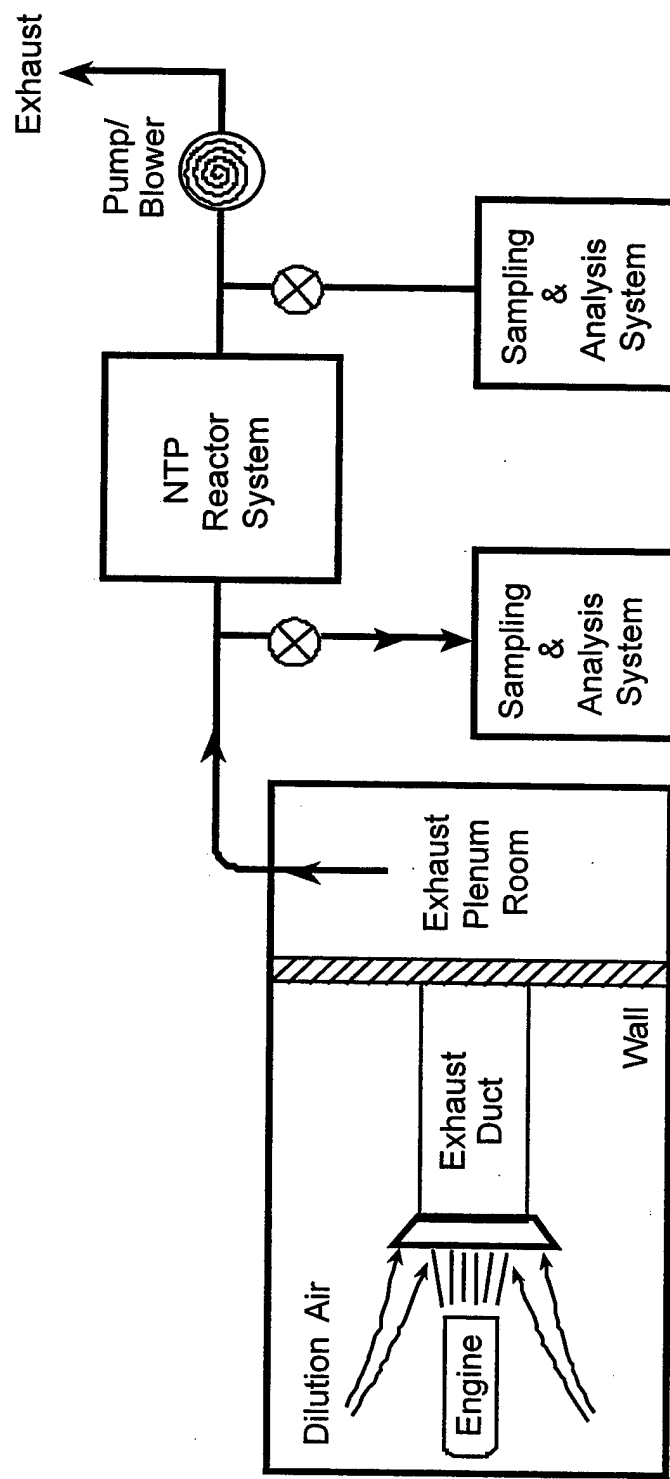
$$(4) \quad \gamma = \frac{\bar{E}}{[X]_0 - [X]} = \frac{-\ln\left(\frac{[X]}{[X]_0}\right)}{G\left(1 - \frac{[X]}{[X]_0}\right)} \quad (= 1/G \text{ at optimum; i.e., } [X]/[X]_0 \sim 1).$$

Scaling laws for pollutant removal (graphical forms)



Energy costs and degree of removal for NO removal in an NO-air mixture with one e-fold plasma specific energies of 50 J/liter and 20 J/liter. NO is a compound whose β -value is not very sensitive to the initial concentration; therefore, the removal energy costs decrease as the concentration increases (note 200 ppm case vs 100 ppm case).

NTP pilot-unit setup for de-NOx field tests



Expected exhaust-gas data and conditions

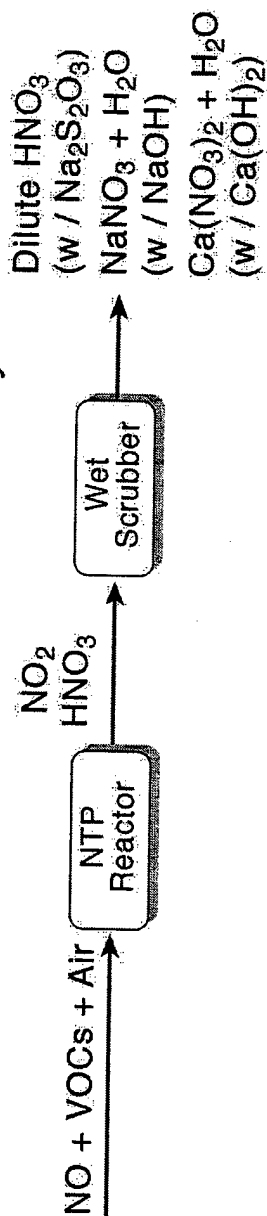
Source Data	Variable	Units	JETC Values	CMTC Values
Gas Flow	Q _{gas}	Nm ³ /h	1.0E+05 1.7E+06	6.6E+03 1.3E+04
Fuel			JP-8	JP-10
Final Exhaust-Gas Composition				
N ₂	CN ₂	%	80.98	78.00
O ₂	CO ₂	%	18.00	21.00
CO ₂	CCO ₂	%	0.50	0.50
H ₂ O	CH ₂ O	%	0.50	0.50
Density (Normal)	D _{gas}	kg/Nm ³	1.283	1.283
Exhaust Gas Temperature	T _{gas}	C	25	25-75
NTP Inlet Temperature	T _{NTPIn}	C	25	25-30
Pressure	P _{gas}	mm Hg	720	720
Emission Data				
NO _x	CNO _x	ppm	36.00	< 10
SO ₂	CSO ₂	ppm	4.59	~ 1
HC (VOC)	CHC	ppm	60.00	20-25
CO	CCO	ppm	53.36	~ 5
Particles	C _{part}	mg/Nm ³	-	~> 1.0
NH ₃ Stoichiometric Ratio to NO and SO ₂ : 1.5 for both JETCs and CMTCs				

NTP de-NOx System Examples

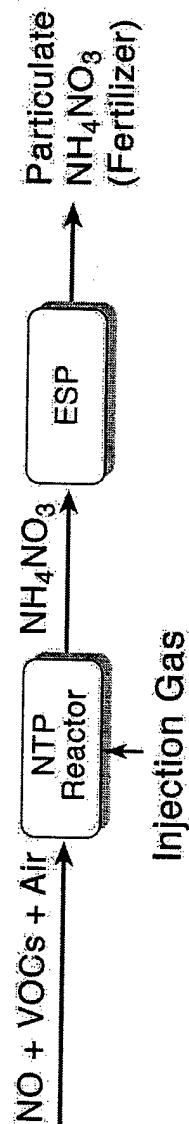
Stand-Alone System



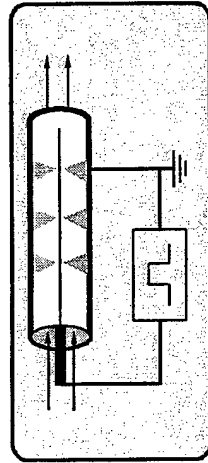
NO → NO₂ Conversion + Scrubber System



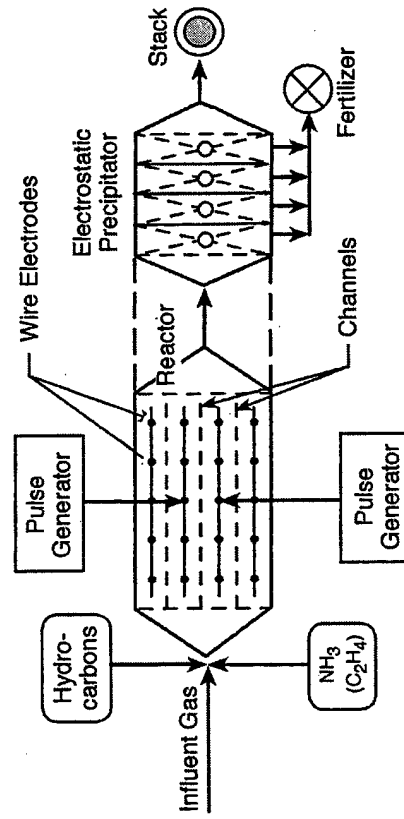
Injection Systems



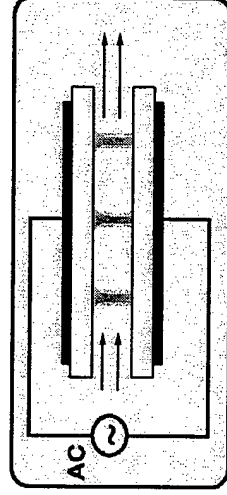
Pulsed Corona System



Pulsed or DC corona



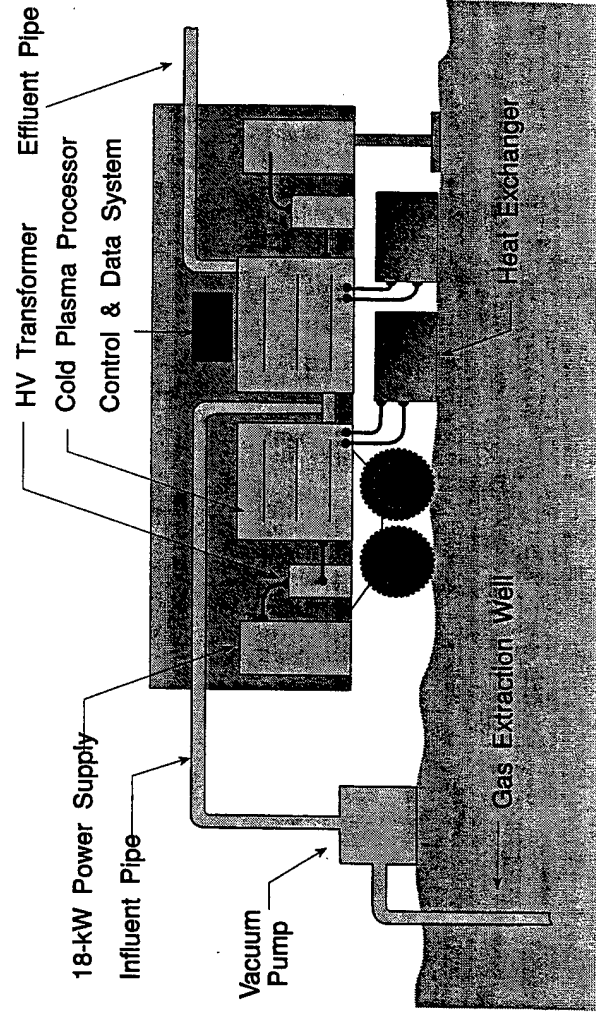
Dielectric-Barrier (Silent Discharge) System



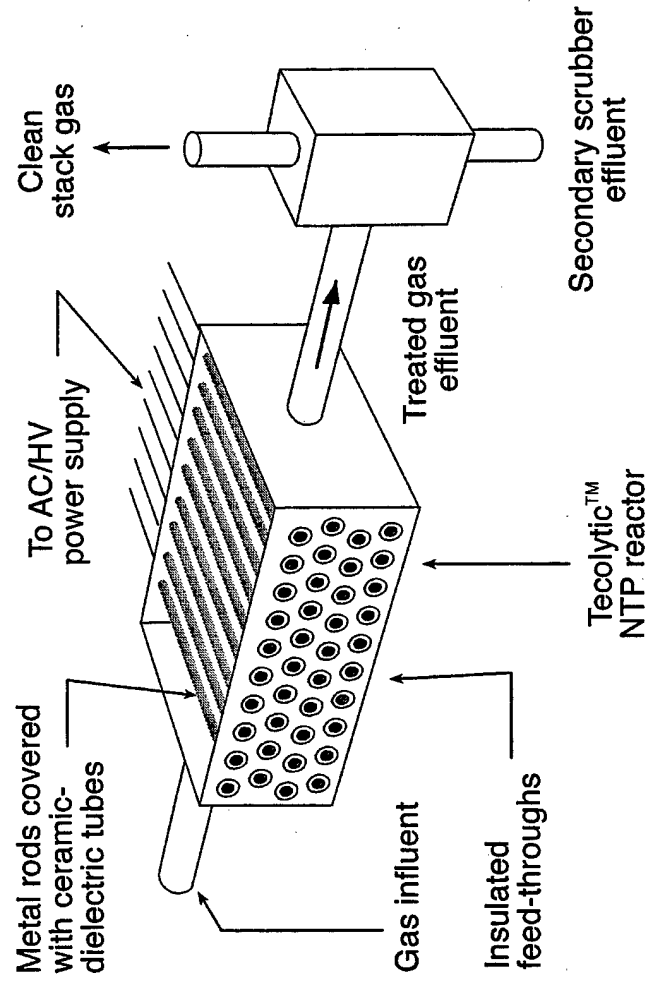
**Silent discharge
(dielectric-barrier discharge)**

Illustration of mobile dielectric-barrier NTP reactor system employed for VOC decomposition tests at McClellan AFB. Each plasma reactor tank operated at up to 10 kW of plasma power.

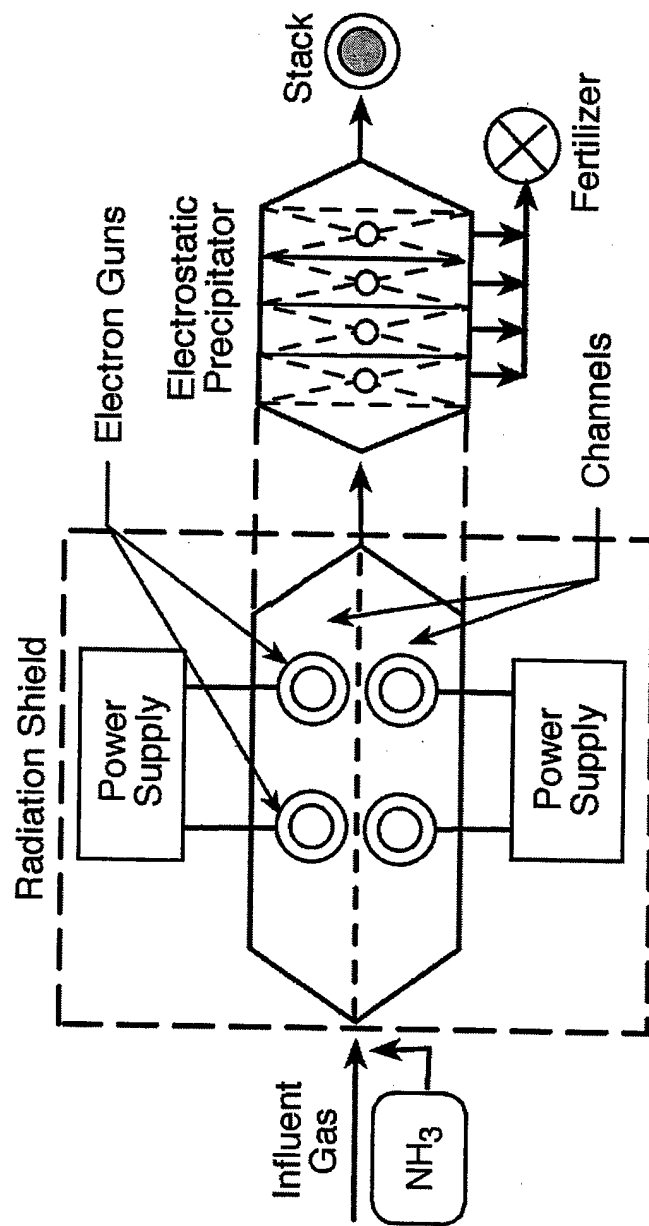
CRADAs with the Electric Power Research Institute (EPRI) & High Mesa Technologies (HMT) were an essential part of the development & fielding of this equipment.



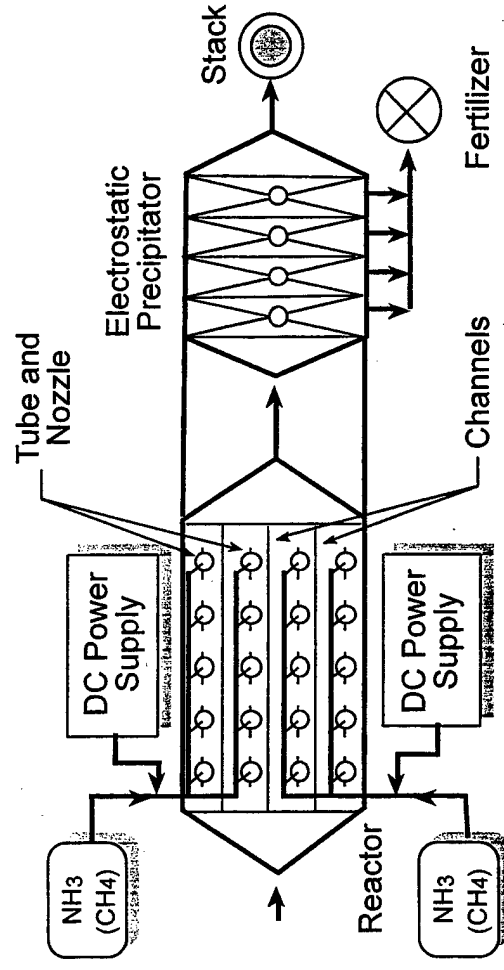
Schematic diagram of commercial Tecolytic™ modified dielectric-barrier NTP reactor system for de-NO_x/SO_x (flue-gas treatment)



Electron-Beam System

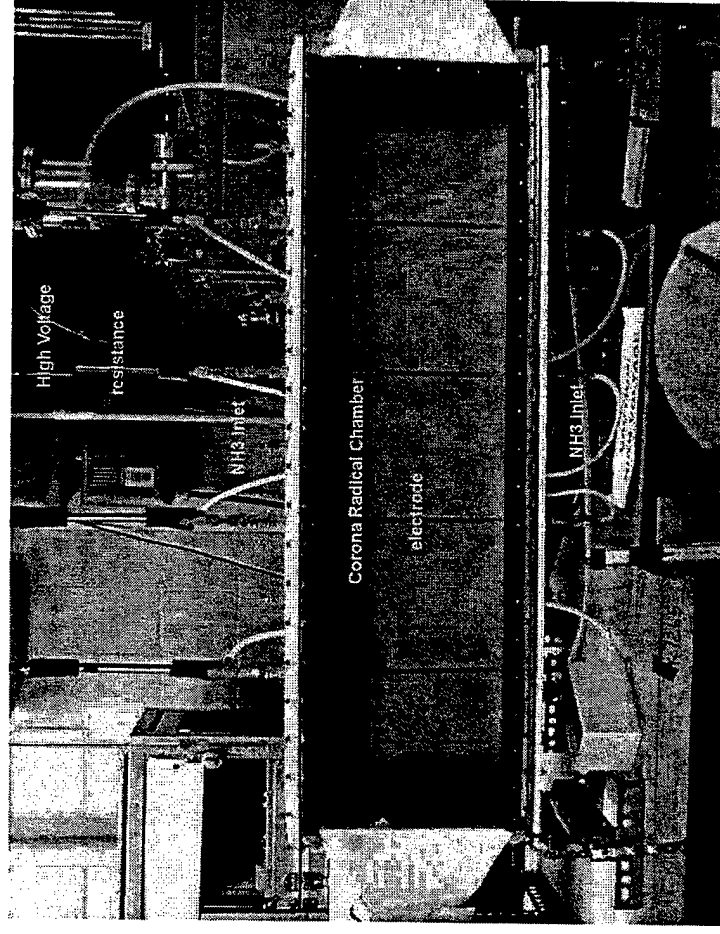
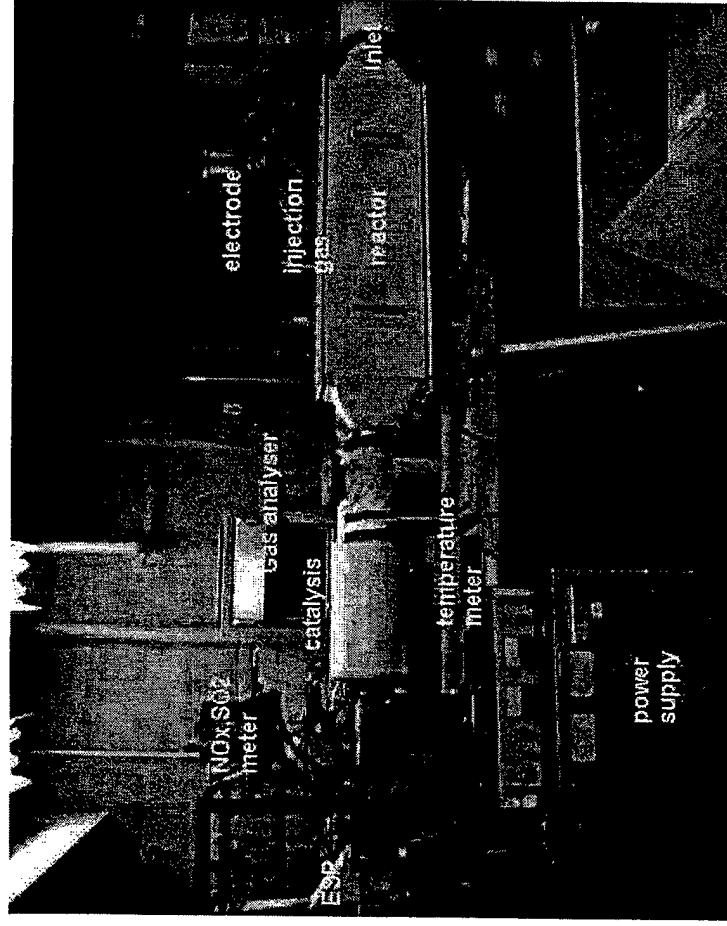


Corona Radical Shower System



Schematic diagram of CRS reactor. Ammonia (NH_3) or methane (CH_4) are added to generate radicals that drive reactions leading to the formation of particulates; these particulates are then captured by the electrostatic precipitator. Some of the captured products are useful for agricultural fertilizer (e.g., ammonium nitrate, NH_4NO_3).

Corona Radical Shower (CRS) Lab-Scale Prototype at McMaster University



We have provided economic analyses for three hybrid NTP systems compared to conventional de-NO_x systems

Conventional	NTP
Selective Catalytic Reduction (SCR) + Wet Scrubbers	Pulsed Corona
Selective Catalytic Reduction (SCR) + Electrostatic Precipitator (ESP)	Electron-Beam
<pre> graph LR IG[Influent Gas] --> WS((Wet Scrubber)) LS[Limestone Slurry] -.-> RMT[Reaction Mix Tank] RMT -.-> WS WS --> RH[Reheater] RH --> SCR((SCR)) SCR --> S((Stack)) </pre>	Corona Radical Shower (CRS)

Example: NO_x-removal power requirement depends on exhaust gas flow rate, characteristic specific energy, and desired degree of removal.

(Assume stand-alone NTP system.)

- **Typical removal scaling:** $[X]/[X]_0 = \exp(-E_s/\beta)$,

where $E_s = P/Q$, plasma power divided by gas flow rate and β = specific energy for one e-fold removal.

For NO, $\beta \sim 10$ J/lit (e-beam); ~ 50 J/lit (electrical discharge).

- **Power requirement for one e-fold removal:** $P = Q \cdot \beta$.

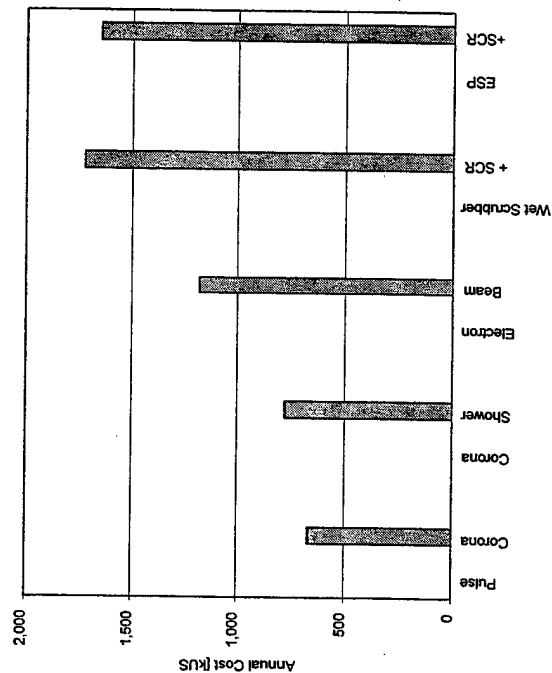
Small Source:	Large Source:
Q ~ 100 SCFM (2832 lit/min)	Q ~ 1×10^6 SCFM (2.83×10^7 lit/min)
P ~ 472 W (e-b), 2.4 kW (e-d)	P ~ 4.7 MW (e-b), 24 MW (e-d)

Benchmarking Basis/Examples for Economic Analyses

	Reactor Type:	Pulsed Corona ¹	Corona Shower ²	Electron Beam ³	Conventional ⁴
	Facility:	JETC Exhaust	'JETC' Exhaust	Auto Tunnel	Power Plant
Parameter	Units	Values			
[NO] ₀	ppm	36	40	10	300
NO _x Removal	%	56	90	70	80
[SO ₂] ₀	ppm	4	4	1	3,050
SO _x Removal	%	90	95	95	90
Energy Yield	g-NO/kW-h	20	17	19	-
Exp. Scale	Nm ³ /h	600	12	40,000	1.9 x 10 ⁶

1) Haythornthwaite et al 1997, 2) Matsuoka et al 1997 (simulated JETC exhaust), 3) Ebara Co. 1998, 4) EPRI 1983, JMIA-EEI 1991.

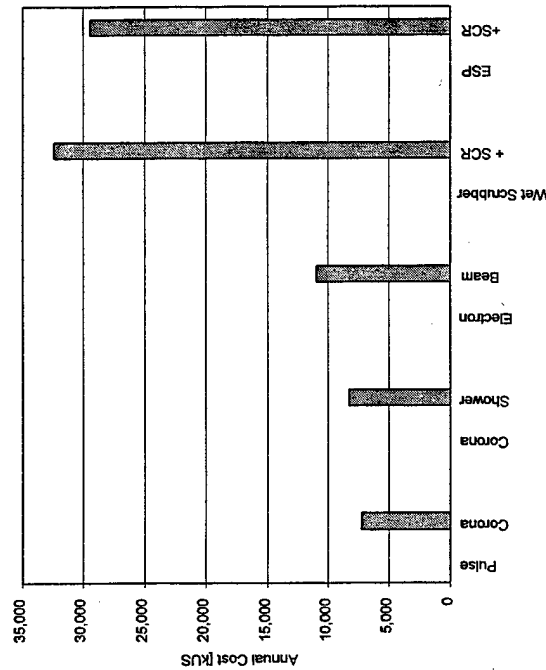
Cost comparisons for various de-NO_x systems (70% de-NO_x, 108 h/wk operation)



Gas Flow Rate: $1.0 \times 10^5 \text{ Nm}^3/\text{h}$ ($5.89 \times 10^4 \text{ SCFM}$)

(k\$)	Pulsed Corona	Corona Shower	Electron Beam	Wet Scrubber + SCR	ESP + SCR
Capital Recovery	345	410	718	36	104
Labor & Maintenance	218	246	377	273	273
Electric Power	99	116	77	123	83
Chemicals & Utilities	6	6	6	1,291	1,192
Total Annual Cost	664	774	1,176	1,723	1,651
Fertilizer Recovery	4	4	4	0	0

Cost comparisons for various de-NO_x systems (cont'd.) (70% de-NO_x, 108 h/wk operation)



Gas Flow Rate: 1.70 x 10⁶ Nm³/h (1.0 x 10⁶ SCFM)

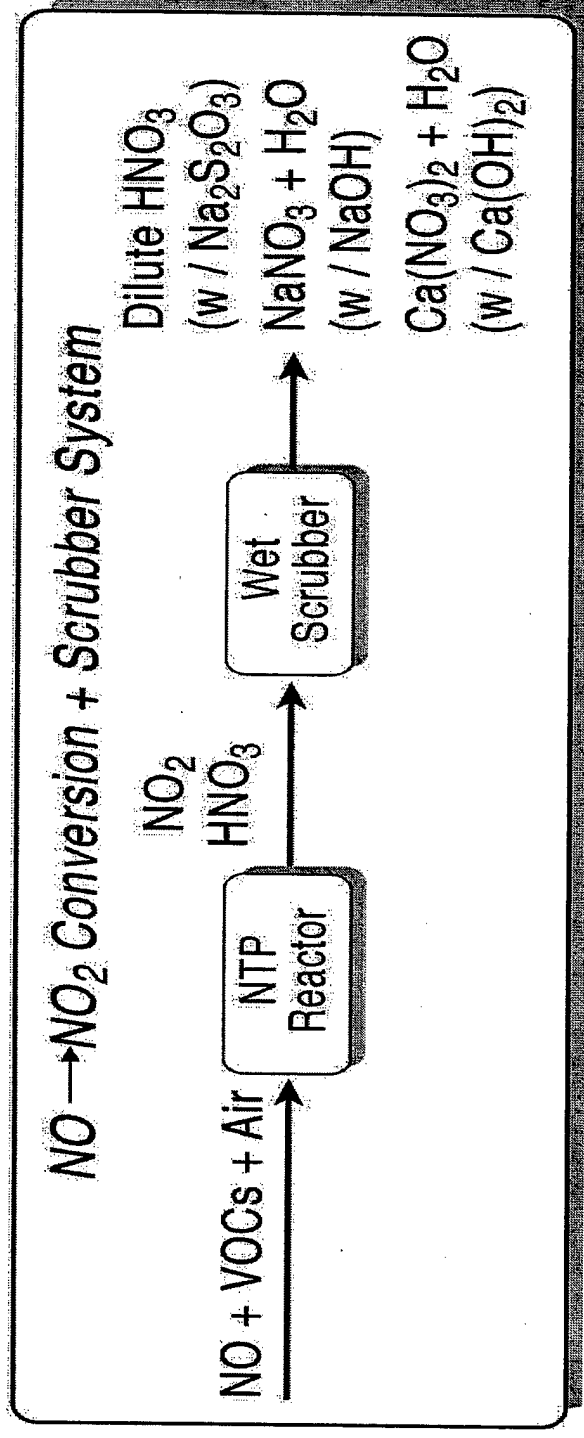
(k\$)	Pulsed Corona	Corona Shower	Electron Beam	Wet Scrubber + SCR	ESP + SCR
Capital Recovery	3,594	4,151	6,504	3,802	3,123
Labor & Maintenance	1,823	2,060	3,061	4,641	4,641
Electric Power	1,674	1,970	1,318	2,082	1,403
Chemicals & Utilities	110	110	110	21,935	20,247
Total Annual Cost	7,139	8,230	10,931	32,459	29,414
Fertilizer Recovery	62	62	62	0	0

Other workers have evaluated two additional NTP de-NO_x & de-SO_x/NO_x systems

Haythornthwaite et al
(Air Force contract)

(Pulsed corona oxidizer + scrubber)

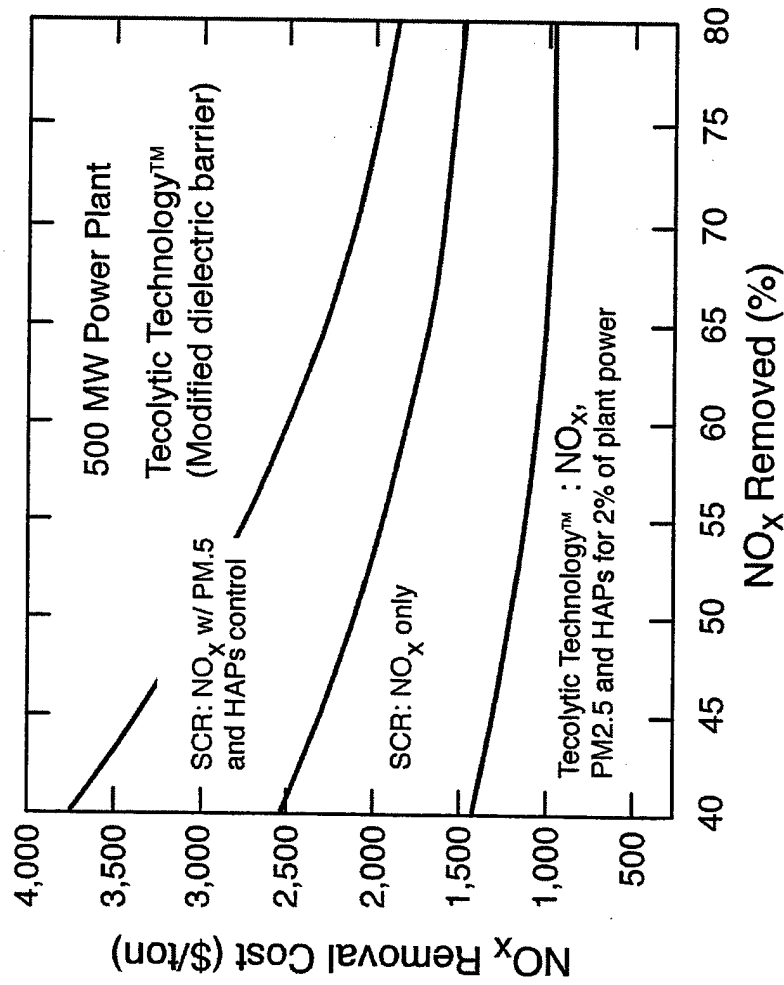
Tecogen/Thermo Power
(Modified dielectric barrier)



**Cost data for
Haythornthwaite et al 1997
4 x 10⁶ SCFM JETC using PCR
NO → NO₂ converter + wet scrubber
(~50% NO_x removal)**

Capital Cost	\$109,200,000	
Capital Recovery	\$14,196,000	
Run Time	10 hr/week	50 hr/week
O & M Costs/Yr	\$523,600	\$2,618,000
Annual Cost	\$14,719,600	\$16,814,000

Cost comparisons for commercial Tecolytic™ modified dielectric-barrier NTP reactor system for de-NO_x/SO_x (flue-gas treatment) show favorable trends



Future trends

Realizing the performance and economic shortcomings of stand-alone NTP reactors, the use of staged or hybrid systems to better match particular air-emissions control applications is being more widely explored

- NTP + injectants
- NTP + catalysts
- NTP + adsorbents

NTP technology may become more common in areas where it has significant advantages over conventional technologies in terms of:

- Increased efficiency and/or economics
- Increased selectivity or throughput
- Better control of final products and/or process
- Unique process streams

Summary

- A greater awareness of the effects of air pollutants (NO_x/SO_x , VOCs, particulates) on our environment and human health has led to more stringent regulations on air emissions.
- Non-thermal plasma technology is being explored as an emissions-control option in several areas (this talk has emphasized NO_x control in jet engine test facilities).
- Hybrid systems (e.g., using injectants) show increased removal yields and favorable economic trends for large-scale de- NO_x systems.
- Rigorous pilot-plant tests are required to provide further data and operating experience to more fully evaluate economic and performance projections.