THE RELATIONSHIP BETWEEN HIGH NITROGEN MUNICIPAL SOLID WASTE COMPONENTS AND NO_x

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ABSTRACT

There is a widely held belief that fuel bound nitrogen is the principal source of incinerator NO_x emissions. This belief leads to the theory that removing high nitrogen components like food and yard and garden waste will reduce NO_x emissions even though incinerators typically release 75 to 90 percent of the fuel bound nitrogen as diatomic nitrogen gas rather than NO_x. This paper explores the validity of the belief using simultaneously collected waste composition and incinerator emissions data, a re-analyses of published incinerator NO_x emissions data, and new uncontrolled NO_x emissions data presentations to confirm that incinerators are staged-combustion devices and NO_x emissions are not related to fuel bound nitrogen levels over the widely observed range. Consequently, while separating compostable materials may be desirable, justifying it on the basis of NO_x minimization is incorrect.

INTRODUCTION

The Waste Analysis, Sampling, Testing and Evaluation [WASTE] Program is an ongoing broad-scope research and development effort designed to develop technical information needed to make environmentally sound solid waste management decisions. The WASTE Program's initial test was conducted at the Burnaby, B.C. Energy-from-Waste [EFW] facility to gather information on the potential relationships between waste components and incinerator emissions and to validate the testing data reduction and data evaluation methodologies to be used in the overall program. The test protocols met EPA Category IV Quality Assurance requirements and were submitted to the project sponsors (i.e., US Environmental Protection Agency [USEPA], Environment Canada, the International Lead Zinc Research Organization [ILZRO] and the Greater Vancouver Regional District [GVRD]) before test execution.

The Burnaby testing was conducted on Unit No. 1 in June 1991. This nominal 240 tonne per day (265 TPD) industrial steam-raising mass burning waterwall [MBWW] municipal waste combustion [MWC] unit began commercial operation in 1988. It is equipped with Martin grates, a two-pass radiant furnace section, followed by a two-pass, parallel flow Sterling boiler, serpentine superheater and economizer. A Flakt APC system, consisting of a flue gas conditioner (spray chamber) to cool the gas stream, a dry sorbent (lime) injection system and a pulse-jet baghouse, provide emissions control. It should be noted that sodium sulfide mercury control reagent was added to the flue gas conditioner during some tests. This unit is not equipped with post combustion NO_x emissions control; rather NO_x emissions are controlled via the inherent staged combustion feature of MBWW MWCs.

The Burnaby tests, consisting of ten 4-hour sampling runs, were conducted the last week of June 1991. The solid waste entering the incinerator was simultaneously sampled and sorted while samples of the solid and gaseous streams leaving the combustor, the dry lime injection chamber and the pulse-jet fabric filter were collected and analyzed. Stack emissions, residues and process parameters were sampled and monitored throughout each run period. Continuous HCl, SO_2, NO_x, carbon monoxide, and total hydrocarbons emissions data were acquired using the plant CEMS system. The facility CEMS was operated in conformance with 40 CFR 60, Appendix F quality assurance requirements. During the first nine runs, 1 to 5 nominal 100 kg solid waste samples per run were sorted into 168 categories.

Process and CEMS data acquisition continued outside the run periods to determine if the plant was operating under normal conditions during testing. Analysis of this data demonstrated that all testing was completed under normal operating conditions.

The result of the Burnaby test program is a unique data set that can be used to study the relationships between MSW components and composition, and residue and gas stream characteristics. Thus, it can be used to identify potential relationships between specific waste components and emissions such as the widely held belief that separating high nitrogen waste components will reduce NO_x emissions. This belief apparently stems from early studies suggesting a seasonal characteristic to NO_x emissions (CARB, 1984; Hahn & Safaer, 1988; and Radian, 1989).

A number of standard statistical techniques were used to analyze the data. Many of the statistical applications are, however, unique because this is the first time that such a large, simultaneous data set — one which simultaneously characterizes the solid waste supplied to the unit, the residues and stack emissions — was collected. The techniques employed in the original analysis can be used to examine The WASTE Program's Burnaby data for relationships between waste components and oxides of nitrogen emissions. This analysis first considers whether there is a relationship between specific, readily identifiable high nitrogen solid waste components and NO_x emissions at the 95% statistical confidence level. An exploratory analysis of the more general question, whether there are any components which appear to have a significant effect on NO_x emissions, was performed next at the 80% statistical confidence level. Finally, regardless of the definitive or exploratory nature of the statistical analysis employed, the results were checked for reasonableness.

PHENOMENOLOGICAL ANALYSIS OF THE BURNABY TEST DATA

The NO_x emissions measured during the Burnaby test ranged between 250 and 300 PPMdv at 7% O_2. These NO_x concentrations are similar to those seen at other MBWW MWC facilities without Selective Non-Catalytic Reduction [SNCR] NO_x control throughout the United States and Canada. They correspond to a calculated nitrogen content of about 0.08 percent by weight in nominal 5,000 Btu/lb municipal solid waste [MSW] if fuel bound nitrogen is the only source of NO_x and all is converted. This is a low value considering the 0.3 to 1% reported nitrogen content range for MSW. Clearly, much of the fuel bound nitrogen is being emitted as nitrogen gas and is not being converted to NO_x. Moreover, unlike fuel bound chlorine or sulfur which are mostly emitted

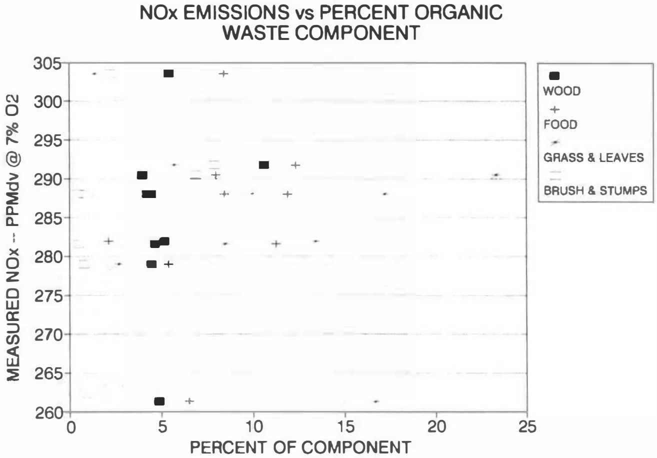

FIG. 1. EFFECT OF WASTE COMPONENTS ON EMITTED NO_x (DATA FROM TABLE 1)

TABLE 1 SUMMARY OF KEY BURNABY TEST NO_x AND READILY IDENTIFIABLE HIGH NITROGEN SOLID WASTE COMPONENTS

RUN	1	2	3	4	5	6	7	8	9
Measured NOx -- PPMdv @ 7% O2	282	292	288	291	304	282	279	261	288
GRASS & LEAVES	8.5%	5.7%	10.0%	23.4%	1.4%	13.4%	2.7%	16.7%	17.2%
BRUSH & STUMPS	0.5%	7.0%	0.4%	6.9%	2.3%	3.1%	0.8%	1.0%	0.9%
WOOD	4.7%	10.6%	4.5%	4.0%	10.0%	5.8%	11.1%	10.2%	11.8%
FOOD WASTE	11.3%	12.4%	11.9%	8.0%	5.5%	5.2%	4.5%	4.9%	4.3%
TOTAL HIGH N ORGANICS*	28.6%	39.6%	29.1%	46.6%	27.7%	29.8%	24.5%	39.3%	42.6%
FUEL NITROGEN	0.41%	0.44%	0.38%	0.66%	0.43%	0.44%	0.34%	0.53%	0.58%
NOx @ 100% Conversion -- PPMdv @ 7% O2	1386	1654	1313	2462	1438	1507	1070	1802	1946

* Includes textiles, rubber and footwear in addition to the above listed organics

as readily identifiable acid gases after they are released during combustion, NO_x emissions come from both the oxidation of nitrogen in the fuel (fuel NO_x) and high temperature oxidation of nitrogen in the air (thermal NO_x). These factors explain why mass balances are not an appropriate way to estimate potential NO_x emission reductions due to changes in fuel nitrogen content.

NO_x emissions are compared to the mass fraction of food, wood, grass and leaves, and brush and stumps found in the MSW during each run in Figure 1. The NO_x emissions and quantity of high nitrogen solid waste components associated with each run are provided in Table 1. Complete detail can be found in The Waste Program's Burnaby Test Report (1993).

Even a casual examination of Figure 1 and Table 1 indicates that there is no apparent relationship between any of the individual high nitrogen components and NO_x emissions. For example, grass and leaves ranged between 2% and 23% by weight of the total waste stream, yet there is no relationship between the NO_x emissions and the amount of grass and leaves. In fact, the NO_x associated with the highest grass and leaves content resides in the middle of the observed emissions distribution and the highest NO_x level corresponds to the lowest observed amount of grass and leaves. Similar statements can be made for wood, food, brush and stumps.

The amount of NO_x which would be produced if all of the nitrogen in the sorted solid waste were converted to nitrogen dioxide (stoichiometric fuel NO_x) was calculated for the observed compositions using available component ultimate analyses and plotted against measured NO_x emissions in Figure 2. A review of Figure 2 reveals that the lowest NO_x emissions occurred simultaneously with a mid-range waste composition (Run 8) as did the highest emissions (Run 5). Identical NO_x levels were found for Runs 2 and 4 which have average and the highest amount of fuel nitrogen, lawn and garden, wood and high nitrogen organic constituents respectively. Looking at extreme values, lowest fuel nitrogen, least amount of high nitrogen waste components (wood, food, grass and leaves, brush and stumps, plus textiles and rubber), the same steam flow rate, excess air level and temperatures during each of the runs, the only variable is MSW composition.

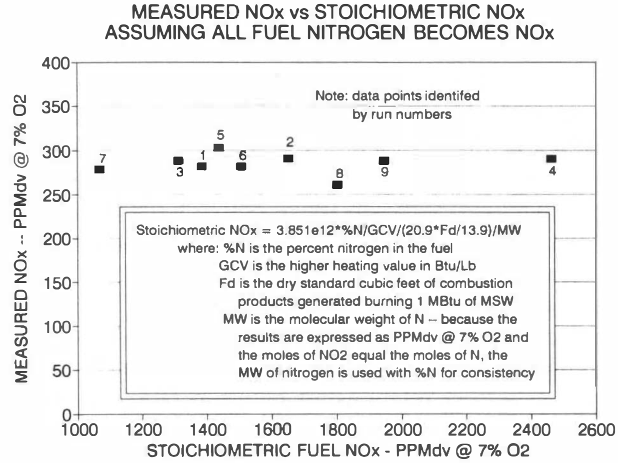

FIG. 2. RELATIONSHIPS BETWEEN EMITTED NO_x AND VARIOUS FUEL PROPERTIES

STATISTICAL ANALYSIS OF THE BURNABY DATA

The slope of the best fit line through the data in Figure 2 is not significantly different than zero. Hence, there is no statistically significant relationship between measured NO_x and stoichiometric fuel NO_x.

Analysis began with a correlation analysis to determine if individual components and emissions were behaving in parallel. The log normal and arcsine distributed sort data were transformed into a normally distributed data set to create a multi-normal distribution to employ standard statistical techniques. This is a key requirement if the statistical confidence levels used to interpret the statistical results are to be accurate. Unlike a simple correlation analysis between two data columns which only compares two parameters, this analysis simultaneously looks at a very large number of correlations and interactions between the 168 sort categories and NO_x to identify parallel behaviour. In order to avoid a large number of false findings (even when working at the 95% confidence level, one out of twenty comparisons—about 700 for the detailed sort categories—will be significant just due to chance alone), critical values of the correlation coefficient were calculated using the Hochberg (1974) GT-2 statistic in lieu of the *t*-statistic.

In examining the correlation coefficients for the identified waste components of grass and leaves, wood, food, and brush and stumps, no significant correlations were found at the 95% confidence level. Even if the analysis is repeated at the lower critical values of r associated with the 80% statistical confidence level typically used in exploratory data analysis, no significance was found. Detailed procedures used to determine the parameters are provided in Volume II, Section 6 of The WASTE Program's Burnaby Test Report (1993).

A broader analysis was performed looking at the correlations between NO_x emissions and all the individual waste sort categories and aggregated component levels (paper, for example, is made up of newsprint, magazines, office waste paper, paper laminates, cardboard, kraft paper, mixed waste paper, etc.). No statistically significant correlations were found at the 80% confidence level.

Following the correlation analysis, the data were re-examined using Factor Analysis. Factor Analysis is a sophisticated mathematical technique that decomposes the correlation matrix using Eigenvalues into a small set of Eigenvectors made of linear combinations of the individual components to fully describe the data (Jambu, 1991). A geometric analogy to a three Factor Analysis is a child's play jack. The individual waste components are described in terms of the fraction of their variability that is accounted for along each axis of the jack. When the factors are studied by rotating the jack about each axis, one at a time, a cloud of data is found in the center with a varying number of points out on the tips of the legs. Those components which are significantly described by an individual factor, rather than being related to the overall behavior of the total data cloud, reside near the tips of the legs. Things found together at the same tip behave in the same manner. Those found outside the cloud, but at the other end of the axis, behave in an opposing fashion.

A slightly different way to look at it is that if a component and a response are co-located at one of the factor tips and they are both statistically significantly different than zero, then there is potential for the component to be causing the response. Caution is required, however, because there is always the possibility that a statistical relationship is merely coincidental. Statistics identify things that are behaving together; either a technical assessment to determine that causality is plausible or finding the same relationship in a number of independent tests is needed to prove that a relationship is more than chance.

Each of the coefficients associated with the Eigenvector (factor) and individual components can be viewed as a new multiple correlation coefficient. Significance coefficients on the factors can then be identified by treating the coefficients like they are multiple correlation coefficients using the GT-2 multiple means statistic in lieu of the Student-*t* when determining the critical value of the correlation coefficient to use in each test.

NO_x emissions were compared to major waste categories. No relationship was found between the NO_x levels and any of the major waste categories using Factor Analysis. The Factor Analysis grouped NO_x with $-\frac{1}{2}''$ fines, but at the opposite end of the factor vector. This implies that increasing fines reduces NO_x emissions. As a result, we conclude that there is no statistically significant potentially causative relationship between NO_x and major waste categories (glass, household hazardous waste, organics, plastics, pulp and paper, small appliances, inerts and OBW).

A similar finding is reached when NO_x emissions are factored against the detailed sort categories frequently employed (for example, pulp and paper is broken down into newsprint, books, magazines, laminates, corrugate and kraft). When the analysis is performed using the most detailed information we have on organic components (paper, plastic, and other organics categories with specific identification of the readily identifiable high nitrogen components) while using major subcategories and groupings for the balance of the waste, the Factor Analysis again indicates that there is no statistically significant relationship between NO_x emissions and any waste category at either the 80 or 95 percent statistical confidence level.

Because of the total number of detailed component categories into which the waste was sorted, computer limitations precluded the simultaneous assessment of the relationship between NO_x and each of these detailed categories in a single computer run. Instead, the detailed categories were split into three groups and the Factor Analysis performed. Again, there is no statistically significant relationship between NO_x emissions and any individual waste component.

TECHNICAL CREDIBILITY ASSESSMENT

Both the observational and statistical analyses of The WASTE Program's Burnaby data found no relationship between fuel nitrogen content and NO_x emissions for mass burning waterwall incinerators over the range of fuel nitrogen values, yard wastes, food, wood, etc. encountered. This finding supports the CARB (1984) conclusion that "... source separation of foodwastes and yardwaste appear to offer the highest potential for *fuel nitrogen* reductions. *Reductions in the NO_x emissions rate due to source separation are probably not significant.*" [emphasis added] However, this conclusion appears contrary to subsequent studies like the "Municipal Waste Combustors-Background Information for Proposed Standards: Controlled NO_x Emissions" (Radian, 1989, p. 2–7) in which limited NO_x emissions data were used to hypothesize that "the observed higher NO_x emissions from massburn/waterwall units during the summer months *may* be due to higher nitrogen content of the fuel because the raw refuse contains more yard wastes, which have a high nitrogen content" [emphasis added].

The Radian discussion begins by referencing the 1984 CARB report to establish that oxides of nitrogen (NO_x) can be formed either as a result of burning the nitrogen in the fuel (called fuel NO_x) or as a result of reaching high enough temperatures that the nitrogen in the air combines with oxygen to form thermal NO_x at higher temperatures. Radian then states that 75% to 80% of the NO_x formed during normal operation of municipal waste combustors is associated with fuel bound nitrogen and references the 1984 CARB report.

The 1984 CARB report does not explain how the 75–80% fuel NO_x contribution conclusion was reached; however, a review of the section on "Fuel NO_x Formation" indicates that the primary reference is Turner, et al. (1972). Turner observes that by increasing flue gas recirculation rates to premixed combustors, thermally derived NO_x is minimized and, after thermal NO_x is minimized, the remaining NO_x can be attributed to fuel bound nitrogen. The CARB report includes a copy of Figure 3 which first appeared as Figure 8 in N. Hirayama's 1975 assessment of the effect of flue gas recirculation on NO_x emissions at the mass burn refractory wall Kita Plant near Tokyo. Based on his data, Professor Hirayama concludes that flue gas recirculation [FGR] decreases NO_x by approximately 25 percent. Using Turner's procedure, the data points plotted in Figure 3 indicate a 75 to 80 percent range for the NO_x potentially attributable to fuel bound NO_x. Importantly, CARB does not conclude that 80% of the nitrogen in the fuel is converted into NO_x. Rather, CARB concludes that 75 to 80 percent of the NO_x being observed is attributable to fuel NO_x formation and 20 to 25 percent is thermal in origin assuming the premixed burner analogy is applicable.

Since the 1975 Hirayama work was done on a refractory wall incinerator, the facility was not operating under the low excess air, high temperature conditions of today's modern MBWW. Consequently, the 20–25% thermal NO_x estimate based on a premixed burner analogy probably does not describe the partitioning (thermal/fuel) of NO_x emissions from modern staged combustion MBWW installations. The Hirayama estimate cannot be updated since the only modern North American MBWW equipped with FGR is the Southeast Resource Recovery Facility [SERRF] in Long Beach, CA and testing without the simultaneous use of SNCR is not permitted. Also, because there is considerable nitrogen in the first stage combustion air supplied in an FGR equipped unit, the only definitive way to establish thermal/fuel NO_x sourcing would be to experimentally determine NO_x emissions while using a nitrogen free oxidant. Because oxidant replacement can only be practically done in laboratory scale apparatus, direct

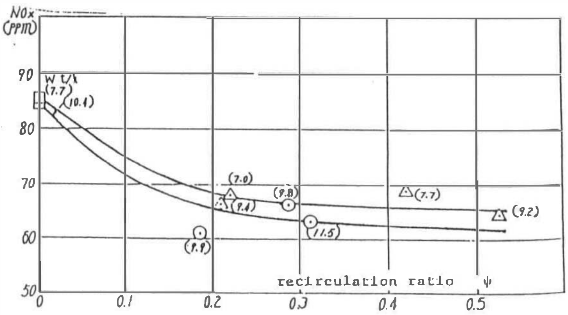

FIG. 3. EFFECT OF FLUE GAS RECIRCULATION NO_x AT THE KITA MBRW INCINERATOR PLANT

partitioning measurements in full scale MBWW MWCs are not realistic and the fuel NO_x contribution can only be inferred using unvalidated, kinetic models.

The central question is whether or not a premixed burner analogy is even applicable and if it is, does it appropriately characterize modern MSW incinerators. In a premixed burner (such as used by Turner, et al.), all of the oxygen needed to complete the combustion process and provide the requisite excess air is carefully and completely blended with the fuel prior to start of combustion. In this case, the nitrogen that is in the fuel is first gassified and sufficient oxygen is immediately available to complete the combustion process and form oxides of nitrogen (as discussed later). In staged combustion, like modern MBWW MWC practice which distributes the air below the grate and through overfire air jets, enough oxygen is provided with the fuel to volatilize the fuel nitrogen, but not enough to complete com bustion and form NO_x. Chemical equilibrium and kinetic considerations indicate that a number of nitrogen enriched intermediaries are formed during the first combustion stage and during the elapsed time before more oxygen is added, these intermediaries react to form gaseous nitrogen. When additional oxygen is introduced in the second stage, the combustion process is completed. But, the second stage only has a very small portion of the fuel bound nitrogen available to burn and form NO_x; the balance of the fuel nitrogen has become nitrogen gas (N_2) which makes up 78 percent of the air we breathe. A substantial portion of the fuel bound nitrogen is converted to N_2, rather than being emitted in the form of the gaseous pollutant, NO_x, in staged combustors like modern MBWW MWCs.

This assessment of two stage combustion is supported by controlled tests using fossil fuels. For example, in the study by Chen, et al. (1982) referenced in Seeker, et al. (1987), the amount of NO_x evolved from pulverized coal in a premixed combustor — where the nitrogen in the oxidant has been replaced by argon to prevent thermal NO_x formation from the air — is between 900 and 1400 ppm NO_x corrected to 5% O_2. When Chen used a *staged combustion system with air as the oxidant*, the same coals formed between 200 and 350 ppm of thermal and fuel bound NO_x.

Work by Habelt (1977) demonstrates similar behavior in full scale tangential, coal-fired boilers operated as staged combustors. Habelt found that only a small percentage of the nitrogen in coal is converted to NO_x and the extent of fuel nitrogen conversion is a function of the oxygen to nitrogen ratio in the fuel itself. Since diverting high nitrogen components increases the oxygen to nitrogen ratio in the remaining solid waste, Habelt's work indicates that there may be an offsetting effect to nitrogen diversion. No net NO_x reduction due to high fuel nitrogen component diversions is plausible.

The preceding discussion explains why *no relationship was observed between fuel nitrogen and NO_x emissions for MSW combustors in The WASTE Program's Burnaby data.* Since the Burnaby facility is a staged combustor, only a small fraction of the fuel bound nitrogen is converted to NO_x. Also, as the nitrogen content increases, the oxygen to nitrogen ratio decreases so less of the fuel bound nitrogen is likely to convert.

Others (Radian, 1989, for example) have speculated that since yard and garden waste is high in nitrogen and exhibits significant seasonal changes, NO_x emissions should exhibit an annual profile that is similar to the changes induced by yard and garden waste disposal if fuel nitrogen content significantly affects MWC NO_x emissions. To test this theory, we first followed the Radian approach and used the results of 116 individual NO_x compliance tests (compliance tests as opposed to CEMS data) for modern, mass burning waterwall incinerators located in North America. While such an analysis cannot explicitly address effects of detailed design and operating philosophy differences, it can identify if the supposition appears reasonable because design and operating differences should become less important as a larger data set is used.

Figure 4 is a normal probability (quantile-quantile) plot of the NO_x data. The data are generally normally distributed and there appear to be a few low data points. These low points are not precisely on the normal distribution line, however, they are close enough that the normal distribution describes the data. That is, the data all seem to come from one family regardless of the testing date or location. When this data is plotted as in Figure 5 to display NO_x emissions versus month of testing, there is no month-to-month difference in NO_x emissions.

The lack of an observed affect was confirmed by comparing monthly box plots of the data as shown in Figure 6. In a box plot, the median of each month's data is identified by an asterisk, the box includes the middle 50 percent (half) of the data and the bars go out to the high and low values as long as they are located within two box lengths (about 1.5 standard deviations) of the median. Outside two box lengths, the data can be considered extreme values and outliers.

A statistically significant difference (95 percent confidence level) between April and May and the rest of the year is found using the graphical comparison technique

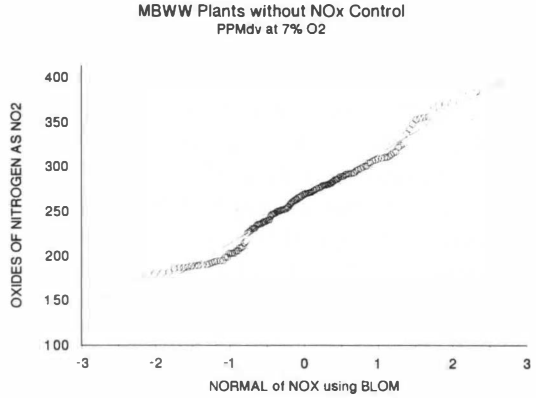

FIG. 4. NORMAL PROBABILITY PLOT OF THE NO_x EMISSIONS FROM NORTH AMERICAN MBWW WASTE-TO-ENERGY PLANTS WITHOUT SNCR

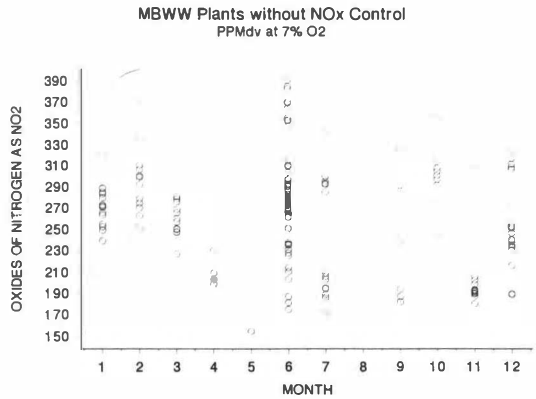

FIG. 5. MONTHLY PLOTS OF THE NO_x EMISSIONS FROM NORTH AMERICAN MBWW WASTE-TO-ENERGY PLANTS WITHOUT SNCR

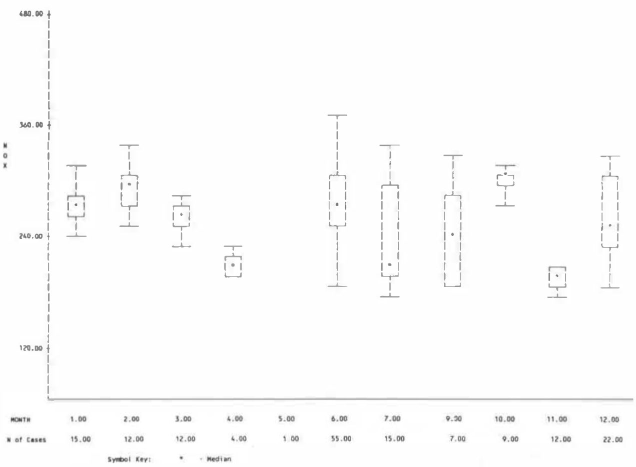

FIG. 6. BOX PLOT COMPARING NO_x EMISSIONS TAKEN THROUGHOUT THE YEAR AT NORTH AMERICAN MBWW WASTE-TO-ENERGY PLANTS WITHOUT SNCR

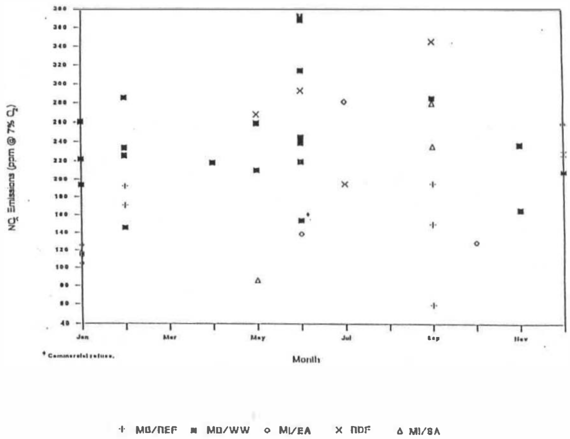

FIG. 7. PLOT OF NO_x EMISSION BY MONTH REPRODUCED FROM THE RADIAN BID FOR CONTROL OF NO_x EMISSIONS

developed by Gabriel (1972) as extended and described in The WASTE Program's Burnaby Test Report (1993). Less NO_X was apparently generated at the beginning of spring clean-up and the growth season than the rest of the year. The implication is that removing yard and garden waste increases NO_X emissions. Since both April and May have few data points, this may be a statistical aberration that would go away if more data were available. If the phenomena is real, it could easily be a consequence of the extra moisture in the yard waste decreasing the peak furnace temperature or shifting the first stage intermediates to produce more reducing species.

This finding is consistent with the data reported by others. An examination of the mass burning waterwall NO_X data plot in the Background Information Document (Radian, 1989) — reproduced as Figure 7 — shows data ranges that do not support a seasonality conclusion. June has the largest range and all other months' data fit within its limits. Even if the June low point is discarded as atypical, the other months' ranges overlap the June results. So, these data do not support the seasonality hypothesis. When each combustor type is separately examined, the data still appear to come from a single distribution. Our conclusion is that there is no seasonal effect and seasonality cannot be demonstrated using the Radian data sets. It is also consistent with an analysis of multi-year NO_X CEMS results (Stultz, et al., 1993).

CONCLUSIONS

Because MBWW MWCs utilize staged combustion where a portion of the combustion air is introduced under the grate with the fuel and the balance down stream of the initial burning zone as secondary or overfire air, this lack of relationship between high nitrogen solid waste components and mass burning waterwall NO_x emissions appears reasonable. A review of the underlying theory indicates that in staged combustion systems, the nitrogen in the fuel is volatilized in the first stage and a significant portion of it is reduced to nitrogen gas before more oxygen is added in the second stage. When the additional air is introduced through the overfire air jets to complete the combustion, the remaining organic nitrogen oxidizes to form fuel NO_x and some thermal NO_x is formed as well.

Over the range of materials combusted during the Burnaby test (from 8% to 38% by weight yard and garden and wood waste and in total between 24% and 47% high nitrogen organics), the fuel nitrogen content did not affect the emissions of oxides of nitrogen. Over the tested range, from .34% to .66% fuel nitrogen content, no discernable affect of fuel bound nitrogen was observed.

There is no causal relationship between high nitrogen solid waste components and NO_x emission. This finding is consistent with the expected performance of a staged-combustion system.

DISCLAIMER

This supplemental data analysis has not been reviewed by the Burnaby test sponsors or all members of the consortium and should be taken as the authors' professional engineering opinions; no official endorsement by the general sponsors should be inferred at this time.

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