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# GLOBAL COMBUSTION SOURCES OF NITROUS OXIDE EMISSIONS

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# ABSTRACT

The existing international knowledge base of emissions and measurements of $N_2O$ from combustion sources is summarized and evaluated in this report based on the available documentation. The emphasis is on identifying problems with existing data and on identifying the areas of greatest uncertainty. The majority of recent experimental results offers consistently low $N_2O$ emission results for combustion energy sources, with the exception of fluidized bed combustors. There is also substantial evidence that some $NO_x$-control methods may contribute to increased $N_2O$ emissions, especially in certain temperature ranges. The report uses the available data to calculate preliminary emission factors which will be used later to estimate the global contribution from these sources to atmospheric $N_2O$.

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## Section 1

## INTRODUCTION

Nitrous oxide has been implicated as both a greenhouse gas and a contributor to stratospheric ozone depletion. Evidence indicates that the concentration of $N_2O$ in the atmosphere has been increasing since the onset of industrialization. Much recent work has been done to characterize the sources and sinks of atmospheric $N_2O$. Until about 3 years ago, between 20% and 30% of anthropogenic $N_2O$ in the atmosphere was attributed to combustion sources. (8, 50, 177) With the discovery of sampling problems, significant uncertainty has been created regarding the contributions of various sources to overall $N_2O$ emissions. This study addresses the relative contributions of various $N_2O$ emission sources and assesses the reliability of emission estimates reported to date.

The purpose of this report is to summarize the available data regarding combustion sources of $N_2O$. The focus is on sources used for energy production, as they have been the cynosure of $N_2O$ emission investigations thus far. From it, areas of needed research may be identified and pursued so that a clearer picture of the contribution of particular sources can be obtained. Without such information, attempts to limit $N_2O$ emissions may result in inefficient outcomes by targetting minor sources rather than the primary causes of the $N_2O$ increase. A survey of accessible literature was conducted and $N_2O$ concentrations from a full range of combustion sources were tabulated. Emission factors were also developed for each category of combustion sources, based on the existing data. Section 2 describes the role of $N_2O$ in atmospheric reactions and discusses the contributions of natural and anthropogenic sources to the overall $N_2O$ inventory. Combustion sources of $N_2O$ emissions are discussed in Section 3, including a summary of emissions measurement data and presentation of $N_2O$ emission factors.

# Section 2

# $N_2O$ IN THE ATMOSPHERE

## IMPACTS AND HISTORY OF ATMOSPHERIC $N_2O$

Nitrous oxide, with an atmospheric concentration measured to be between 310 and 330 ppb, is the second most abundant nitrogen species in the atmosphere, less prevalent than only the elemental $N_2$ which comprises over three-fourths of the atmosphere. (156, 43) Evidence indicates that the concentration of $N_2O$ has been increasing since industrialization, presumably because of the addition of anthropogenic sources to the existing natural sources. The increase has been observed only recently. Those studying the increase still do not understand fully or agree on its causes or its overall importance to global climate change. However, $N_2O$ has been implicated as both a greenhouse gas and a contributor to stratospheric ozone depletion.

Ozone is located primarily in the stratosphere (approximately 90%) with the remaining 10% in the troposphere. Since ozone is the only atmospheric gas that absorbs energy in the ultraviolet (UV) wavelengths, decreases in the atmospheric ozone concentration will result in increased amounts of solar UV radiation reaching the earth's surface. In fact, studies indicate that the percentage increase in UV radiation reaching the surface corresponds to approximately twice the percentage decrease in the ozone concentration (156); that is, a 5% decrease in atmospheric ozone concentration would result in an increase of approximately 10% in the UV radiation reaching the earth's surface. This increase would lead to a corresponding rise in the global mean temperature.

Such decreases in the concentration of ozone are caused in part by the presence of nitric oxide (NO) which facilitates the conversion of $O_3$ to $O_2$. The only source of NO formation in the stratosphere is $N_2O$, via the reaction

$$N_2O + O \dashrightarrow 2NO$$

Thus, it is estimated that 60 to 70% of global ozone destruction is the indirect result of $N_2O$. (<u>156</u>) It is estimated that a doubling of the $N_2O$ concentration would result in a 12% decrease in atmospheric ozone levels. (<u>43</u>)

$N_2O$ also contributes to climate change because it is a greenhouse gas. It is estimated that $N_2O$ may be responsible for between 5% and 6% of the anthropogenic greenhouse effect. (<u>79</u>, <u>154</u>) Greenhouse gases trap surface heat that is reradiated in the infrared (IR) wavelengths, thus reducing one of the mechanisms of heat loss and affecting the overall heat balance. (<u>92</u>) The majority of the greenhouse effect is caused by water and carbon dioxide.

The concern over increased atmospheric $N_2O$ concentration is also based on its long decay time of about 150 years, the longest of any greenhouse gas. (<u>154</u>) Thus, changes in the concentration of $N_2O$ may have particularly long-term repercussions, the extent of which are not yet known.

In the past 100 years, the atmospheric $N_2O$ concentration is estimated to have increased by about 20 ppb. (<u>73</u>) The rise has been estimated to be between 0.2 and 0.3% annually. (<u>112</u>, <u>43</u>, <u>156</u>, <u>73</u>) This is supported by evidence that, in pre-industrial times, the $N_2O$ concentration was 280-290 ppb. In the 1960s, it rose to 290-295 ppb; it now exceeds 300 ppb. (<u>73</u>) It is estimated that, by the year 2050, the $N_2O$ concentration will be as high as 360 ppb. (<u>73</u>)

## CHARACTERIZATION OF $N_2O$ EMISSION SOURCES

The relatively recent nature of the increase in atmospheric $N_2O$ concentration suggests that industrialization-related human activities are responsible for the change. Natural sources of $N_2O$, such as microbial nitrification, denitrification in soils and ocean surface water, lightning, and natural forest fires, are assumed to have remained approximately constant over the centuries. The sources of increasing $N_2O$ are suspected to be fertilizer use, biomass burning, and fossil fuel combustion. In addition, other anthropogenic $N_2O$ emission sources have been identified recently and are being investigated. Global $N_2O$ removal from the stratosphere by photolysis is on the order of 10,000,000 tons per year on a nitrogen basis. The annual growth rate or accumulation is 3,000,000 tons. In other words, sources exceed sinks by 30%. (73)

Several studies have attempted to identify the relative contributions of various sources to global $N_2O$ emissions. Hao (73), Kavanaugh (83), and Crutzen (41) provide examples of such estimates. However, the reliability of such estimates depends on the emission factors used in their calculations, as well as on the accuracy of assumptions involving human activities. In several cases, $N_2O$ emissions were estimated based on reported $NO_x$ emissions; it appears now that the assumed proportionality between $N_2O$ and $NO_x$ may be unjustified. Thus, it should be noted that Hao's estimate tends to overemphasize the importance of combustion sources. Nevertheless, such estimates do provide a general idea of the relative contributions of the major combustion sources of $N_2O$ emissions. For instance, Hao's data in Table 2-1 show the distribution by industrial sectors. According to Hao's results, 72% of $N_2O$ combustion emissions are attributable to fossil fuel combustion for electrical generation.

Kavanaugh made predictions for $N_2O$ emissions through the year 2025 based on Hao's estimates. He extrapolated emissions for different regions of the world and emission source categories by making certain assumptions about the path of development, such as that China's industrialization will rely heavily on fossil fuel combustion to provide utilities. Although Kavanaugh's categories are defined differently than Hao's, it is clear from Table 2-2 that China and the rest of the world will contribute the majority of anticipated increases in the atmospheric $N_2O$ concentration with their increased electricity consumption.

The estimates of Hao and Kavanaugh are problematic in that they are based largely on correlations of $N_2O$ emissions to $NO_x$ emissions by generalized

## Table 2-1

## GLOBAL $N_2O$ EMISSION ESTIMATES

| Emission Source | Total $N_2O$ Emitted ($10^6$ tons Nitrogen) |
|---|---|
| Electric Utility | 1.68 |
| Steel Industry | 0.001 |
| Other Industry | 0.49 |
| Household | 0.09 |
| Transportation | 0.07 |
| TOTAL | 2.33 |

Source: Hao et al. 1987 (73).

Table 2-2

GLOBAL $N_2O$ COMBUSTION EMISSIONS BY REGION AND CATEGORY

| Region or Category | 1960 | 1975 | 2000 | 2025 |
|---|---|---|---|---|
| **REGIONS (% of TOTAL)** | | | | |
| U.S.A. | 23 | 24 | 25 | 16 |
| Western Europe | 20 | 17 | 13 | 9 |
| Japan, Australia, New Zealand | 5 | 9 | 8 | 6 |
| Soviet Union | 33 | 30 | 21 | 15 |
| China | 8 | 10 | 15 | 18 |
| Rest of World | 10 | 10 | 18 | 35 |
| **CATEGORIES (% of TOTAL)** | | | | |
| Utility | 55 | 53 | 55 | 54 |
| Industry | 40 | 41 | 41 | 42 |
| Household | 5 | 6 | 4 | 4 |
| Transport | 0 | 0 | 0 | 0 |
| **TOTAL ($10^6$ tons of Nitrogen)** | **2.91** | **4.46** | **9.22** | **13.68** |

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Source: Kavanaugh 1986 (83).

factors for combustion sources, which have recently been called into question. Perhaps a more accurate estimation of emissions for combustion sources is offered by de Soete in Table 2-3. (50) His estimates are based on data collected since 1987 which incorporated measures to avoid the sampling artifact which may have plagued previous data. Table 2-3 summarizes his results. His figures indicate a total contribution from combustion sources about 70% less than Hao's estimates, with a much larger proportion of emissions generated from transportation sources.

The drawback of de Soete's estimate is that current estimates of emissions from all sources do not correlate with the observed increase in global $N_2O$ concentration. New estimates indicate that fossil fuel combustion contributes on the order of 3% of the anthropogenic flux (106), or 10% of the annual $N_2O$ increase, down from about 20% previously estimated. (50) Some natural sources have been determined to be contributing less $N_2O$ than originally believed. For instance, the massive oceanic source proposed by Hahn in 1974 has been disputed by later work. (41) And, discrete phenomena such as lightning are also believed to comprise only minor contributions to atmospheric $N_2O$. Thus, contributions from other known sources must be greater than previously believed or there must be additional sources that have not yet been quantified, such as nylon production. Investigators are now trying to locate the remainder of the sources of the "missing $N_2O$."

There are several known sources of $N_2O$ which are now thought to be more important emitters of $N_2O$ than previously believed and, thus, are the subject of current investigations. Chemical fertilizer, because of its wider-spread application and only partially understood interactions with soil and biota, may be a larger source than previously thought. Recent work has demonstrated that emissions from nylon production could account for as much as one-tenth of the annual increase in atmospheric $N_2O$ levels. (136) Motor vehicle emission estimates (3.5% of total $N_2O$) are greatly dependent on assumptions about the age of catalysts and the usage patterns of vehicles in the world. (43) Catalytic converters have been linked to higher $N_2O$ emission by Swedish studies. (<u>43</u>, <u>50</u>) Non-catalyst cars had $N_2O$ emissions of 0.22% of $NO_x$, which amounted to less than ambient air. Oxidation, dual-bed, and three-way catalysts have been shown to have emissions on the order of 1.8 to 8.2% of

## Table 2-3

## COMBUSTION CONTRIBUTIONS TO GLOBAL $N_2O$ EMISSIONS

| Combustion Source | Contribution to World $N_2O$ Emissions ($10^6$ tons per year) |
|---|---|
| **STATIONARY** | |
| Coal and peat | 0.24 |
| Heavy fuel oil | 0.09 |
| Natural gas | 0.03 |
| **AUTOMOTIVE** | |
| Gasoline | 0.13 |
| Gasoil, diesel | 0.14 |

Source: de Soete 1990 (50).

$NO_x$. Both disabled exhaust gas recirculation and rich idle led to higher $N_2O$ emissions. High mileage cars had almost double the $N_2O$ emissions. (43)

Current estimates of all global sources of $N_2O$ (assuming no fluidized bed combustors and 50% of cars have aged catalysts) predict emissions of 1,260,000 tons per year. (50) Previous estimates suggested values of 2,800,000 tons per year. And the shift toward higher temperatures caused by aged catalysts result in approximately triple the expected emissions (0.11-0.13 g/km for aged, 0.03-0.037 g/km for new). (50)

Perhaps the most significant potential sources of the "missing $N_2O$" are biomass burning, specialized forms of combustion, and indirect reactions of fossil fuel combustion products. The importance of biomass burning seems to be substantiated by recent findings which suggest that biomass burning and related soil emissions due to rapid heating, especially in tropical zones, may contribute more than the 7% which is currently attributed to this source. (41, 32, 177) Emissions from fossil fuels appear to be attributable primarily to processes such as fluidized bed combustion and urea injection (emissions of 20 to 100 ppm and 100 to 200 ppm, respectively) rather than conventional combution sources (emissions of 1 to 10 ppm). (92, 50, 106) Additionally, the NO, $NO_2$, and $SO_2$ released in combustion processes may prove to have indirect links to $N_2O$ formation in the troposphere, accounting for an additional 1.9 Mtpa. (106, 50, 92) Furthermore, although $N_2O$ levels in flue gases per se show them to be only a minor direct contribution to atmospheric $N_2O$, reactions of the varied products of combustion in stack plumes and in the atmosphere may increase the overall contribution to atmospheric $N_2O$. (167)

# COMBUSTION SOURCES OF $N_2O$ EMISSIONS

Research into $N_2O$ emissions from combustion sources has included both theoretical and experimental investigations. Existing work on sources of $N_2O$ from combustion includes substantial efforts to identify the mechanisms involved in $N_2O$ formation and destruction during combustion. Other research efforts have focused on methods of measuring $N_2O$ emissions. Unfortunately, much of this work is now suspect because of the discovery of $N_2O$ formation in gas samples under certain conditions.

## THEORETICAL INVESTIGATIONS

The work of Roby and Bowman (<u>153</u>), and later of Miller and Bowman (<u>123</u>), provides the foundation for characterizing reactions involving $N_2O$. Roby and Bowman proposed the reaction sequence

$$NH + NO \dashrightarrow N_2O + H$$
$$N_2O + H \dashrightarrow N_2 + OH$$

as the primary source of $N_2O$ formation. (<u>153</u>) In addition, a small amount (less than 10%) of $N_2O$ might be formed by the reaction

$$NH_2 + NO \dashrightarrow N_2O + H_2$$

and the following reaction might account for removal of less than 1% of $N_2O$ formed (153):

$$N_2O + M \dashrightarrow N_2 + O + M.$$

De Soete (47) expanded the understanding of the pertinent reactions by taking into account the complex network of overlapping chemical pathways involving the interactions of $O_2$, $NH_3$, $H_2O$, $N_2O$, NO, and $N_2$. The specific steps in the proposed mechanism for $N_2O$ formation and destruction, are (47):

$$NH + NO \dashrightarrow N_2O + H$$
$$CNO + NO \dashrightarrow N_2O + CO$$
$$N_2O + H \dashrightarrow N_2 + OH$$

Miller and Bowman have also proposed mechanisms for $N_2O$ formation from $N_2$, from NCO and NO, and from NH as an intermediate. (123)

De Soete went on to show the effects of catalysts and ammonia on NO formation. He demonstrated that high efficiency metal oxide catalysts for NO decomposition display high production of $N_2O$ within a specific temperature window. In addition, catalytic decomposition of ammonia and bound oxygen gives NO, which reacts with adsorbed ammonia to give $N_2O$. However, in the "optimum temperature window," there is more NO formation (from $O_2$ and adsorbed ammonia). (47) Furthermore, de Soete proposed that fuel staging and non-catalytic selective NO reduction (NCSR) lead to $N_2O$ emission enhancement at low temperatures. $N_2O$ formation results from the interaction of NO and $NH_3$. (47)

In grate-fired combustion systems, $N_2O$ formation is prevented in the furnace center by high temperatures, but $N_2O$ is formed at walls. Grate combustion systems have typical emissions of 1 to 35 ppm. (2) De Soete reported that $N_2O$ formation is effectively reduced above 800°C and $N_2O$ is unstable above 900°C for heterogeneous phases in a quartz reactor. $N_2O$ is also destroyed if there is minimal oxygen (less than 1%) present. (2)

In combustors, the prevalent conditions of temperature and pressure dictate that the expected reaction of

$$N_2O + H \longrightarrow N_2 + OH$$

rapidly destroys $N_2O$ formed early in the combustion process, thus preventing it from reaching the furnace exhaust, according to the work of Lyon, Kramlich and Cole. (112) Nevertheless, it is important to note that this destruction of $N_2O$ may not occur in fluidized bed combustors because of the lower temperatures typical of these systems. (112)

Kramlich observed that, at higher temperatures, NO formation is favored because $N_2O$ destruction is efficient and the precursors to $N_2O$ formation are oxidized. Thus, since $N_2O$ does not tend to survive a fuel-rich environment, staged combustion may yield lower $N_2O$ emissions. (156)

## EMISSION MEASUREMENTS

In 1988, Muzio and Kramlich noted an artifact in the prevalent sampling procedures for measuring $N_2O$ from combustion sources. (130) Lyon, Kramlich, and Cole (112) identified a reaction sequence that is thought to explain this phenomenom in flue gas samples. Under certain conditions, NO in the samples oxidizes to $NO_2$, which, in the presence of sulfite, forms nitrite ions. The latter then form hyponitrite ions which result in $N_2O$. This sequence is thought to be responsible for the sampling artifact which resulted in high $N_2O$ emissions reported in investigations prior to 1987.

The discovery of a sampling artifact calls into question the validity of a large portion of the existing database. However, since that discovery, important work has been done both to develop reliable methods of measuring $N_2O$ concentrations and to begin to rebuild the database for emissions from combustion sources. Still, there are significant gaps and uncertainties in those data.

## Problems with Existing Data

Investigators discovered evidence for the in vitro production of up to 300 ppm $N_2O$ in as little as 5.5 hours. (112) According to Muzio, the identification of the sampling artifact calls into question "all existing data which involved container sampling." (106)

Muzio showed that tests yielded high $N_2O$ levels even when $SO_2$ was added downstream of the flame zone, where combustion had been completed. (133) In effect, the conditions for sample flask formation of $N_2O$ include the presence of condensed water, sulfur dioxide, NO, and low pH resulting from aqueous reactions between $SO_2$ and $NO_x$. However, Cofer et al. reported no statistically significant differences in the $N_2O$ mixing ratios between in situ and 4 to 8 hours analyses. Growth was observed 10 and 21 days after sampling. (32) Furthermore, for automobile emissions, the sampling artifact is not believed to be important as the $SO_2$ levels in such samples are on the order of 1 ppb, which is significantly less than the 500 ppb threshold identified by Muzio for significant in vitro $N_2O$ formation. (43)

Nevertheless, the identification of the sampling artifact does have direct relevance to the work of Hao et al. in 1987. Their work reported a range of $N_2O$ emissions for oil, gas, and coal combustion sources from 2.3 to 204 ppm, from which they derived a generalized emission factor for the ratio of $N_2O$ to $NO_x$ of 0.58 (±40%). (73) In vitro formation of $N_2O$ could account for the anomalously high emissions of 204 ppm and could invalidate the emission factor ratio of 0.58 $N_2O$ to $NO_x$ which they derived.

In light of these considerations, much recent work has been focussed on developing accurate means of measuring $N_2O$ emissions which preclude this sampling artifact. Two approaches have been considered: (1) avoiding the storage of samples by using on-line analysis, and (2) removing one of the components necessary for in vitro $N_2O$ formation.

Montgomery et al. developed and tested an on-line, selective, non-dispersive, infrared analyzer. They compared data obtained from pH-corrected grab samples, normal grab samples, and continuous IR-analyzed samples, finding that the pH-corrected and the continuous IR readings both gave good agreement with low $N_2O$ levels and were not a substantial percentage of the $NO_x$ emitted. (125)

EPRI also developed an on-line analyzer to measure emissions at 14 utility power plants, resulting in reported emissions levels much lower than previous results. The two instances of slightly higher $N_2O$ concentrations are believed to be the result of residues in the sampling lines. The only anomalously high $N_2O$ emissions level is believed to be related to the use of urea at the plant in question. (92)

Linak et al. noted that on-line analysis yielded significantly lower emissions than previously reported. (106) Muzio et al. reported similar results from investigators who tested two means of avoiding the sampling artifact. The first, drying the mixture before storage, partially eliminated the formation of $N_2O$ by removing the aqueous phase. The second method, using sodium carbonate solutions, was found to remove $SO_2$ without impacting the $N_2O$ or $CO_2$ concentrations. (133)

## Valid Test Data

Experiments employing one or more of the methods described above for avoiding in vitro formation of $N_2O$ were used in a variety of investigations. Tables 3-1 through 3-7 summarize those results. For each operation tested, the mode of analysis or sampling procedure is noted. Many of the American investigations rely on continuous analyzers, while the European methods rely primarily on drying the samples with silica gel or condensers.

Previous investigations had found results on the order of $N_2O/NO_x$ ratio of 0.3 to 0.6. (73, 133) Recent results estimate this ratio to be on the order of 0.1 or less. (133) However, there is also substantial evidence which suggests that there is no basis for a correlation between $NO_x$ and $N_2O$ levels. (106)

For the 96 coal-fired boilers tested by the authorities indicated, the $N_2O$ emissions were in the range of 1 to 444 ppm, as indicated in Table 3-1. Although there were some variations among the types of boilers and grades of coal, the average emission factors noted were quite similar, with the exception of the results for fluidized bed combustors. Indeed, this discrepancy is illustrated by the difference in the average emission factors reported in Table 3-8.

Fluidized bed combustors showed significantly higher emissions, as illustrated by the reported average of 123.5 ppm in the 18 tests reported here. These results are in agreement with the $N_2O$ emissions observed by de Soete (50-450 ppm). (2) However, the emission factor of 0.1658 $lb/10^6Btu$ calculated for circulating fluidized bed combustors is slightly lower than the values of 0.2901 to 0.7044 $lb/10^6Btu$ reported by Mjornell et al. (124) These results are not surprising in light of previous work which established that considerable $N_2O$ is formed when combustion occurs below 900°C, which is generally the case in fluidized bed combustion. (2)

For natural gas-fired boilers, the $N_2O$ emissions were significantly lower than those for coal, with a range of 0.26 to 4 ppm. These results are shown in Table 3-2. This result is not unexpected because of the generally "cleaner" nature of natural gas combustion, although it should be noted that the average difference in emissions between coal-fired and natural gas-fired combustors is significantly less than the range of values noted for coal, so that this distinction in emissions between these two fuel sources may not be statistically supportable.

Although the data identified for gas turbines were limited to 2 field tests, these values are only slightly higher than those provided by the work of Andersson and Brannstrom-Norberg. (8) The results were reasonably consistent with a range of 1 to 4 ppm. The emission factor identified for this type of facility is given in Table 3-8.

For oil-fired boilers, the emissions seem to vary greatly depending on the grade of oil used. Table 3-3 summarizes the emissions for light oil-fired combustors, and Table 3-4 provides $N_2O$ emissions data for heavy oil-fired combustors. Whereas for light oils the range reported was from 1 to 40 ppm, emissions as high as 73 ppm were recorded for heavy oils. The emission factors determined for oil-fired systems are included in Table 3-8.

The results for oil-fired combustion with urea injection show significantly higher emissions than for the standard wall-fired combustors. This result is consistent with Muzio's findings (132) which illustrated a distinct increase in $N_2O$ formation with the injection of urea, ammonia, or cyanuric acid in certain temperature ranges.

Results for peat-burning combustors are indicated in Table 3-5. Although the emissions for wall-fired combustors seem higher than for coal at 19.7 ppm, the figures for fluidized bed combustors are significantly lower at 44.8 ppm.

Table 3-6 summarizes the results for combustors fired by other fuels, primarily mixes of peat and coal. With a range of 1 to 69 ppm, these emissions are similar to those reported for coal and peat.

Emissions from combustors fired by biomass, primarily wood, are presented in Table 3-7. The concentrations of $N_2O$ reported are consistently lower than any of the other categories, with an average of 5.3 ppm. However, because of the lower energy content of wood, the emission factor, on a basis of energy produced, is in line with the others noted in Table 3-8.

Although some work has been reported on emissions from open biomass burning in wildfires, those results vary significantly. For instance, Cofer et al. estimated $N_2O$ emissions to be 0.01-0.02% of $CO_2$ emitted. (32) Sloan, conversely, reported a range of 0.02-0.50%, with an average of 0.22%. Although these results do suggest that biomass burning is a large source of $N_2O$, the evidence reported here is not conclusive.

## Gaps in Emissions Data

Although the $N_2O$ emissions data presented here represent a fairly comprehensive database on emissions from combustion sources, they are limited both in the breadth of testing and the degree of accuracy. The number of boilers tested with reliable methods is relatively small, so that the large range of relatively few data points makes generalizing average emission factors tenuous. Particularly important are the fluidized bed combustor data, which, with a range of 0 to 400 ppm, could represent a very important source of emissions. However, the relatively small number of tests would make such conclusions questionable at this point. Furthermore, a significant gap in the data exists for non-energy-producing combustion sources, specifically open biomass burning.

Data for coal-fired boilers are more substantial, although, once again, a range of 0 to 80 ppm makes generalizations difficult. Until further testing yields reproducible results, only rough estimates are possible. In any case, it does seem quite clear from this data that the emissions are much less than reported in previous investigations which did not take into account the sampling artifact.

Table 3-1

## COAL-FIRED COMBUSTOR $N_2O$ EMISSIONS

| Source (Reference) | Analysis Method (Water Removal) | $N_2O$ Content ppm dry, 3% $O_2$ | Boiler Type |
|---|---|---|---|
| **WALL-FIRED** | | | |
| Kokkinos (92) | On-line | 11 | |
| Kokkinos (92) | On-line | 2 | |
| Kokkinos (92) | On-line | 6 | |
| Sloan (167) | NDIR1 | 13 | |
| Sloan (167) | NDIR2 | 9 | |
| Muzio (129) | Continuous IR on-line | 11 | |
| Muzio (129) | Continuous IR on-line | 6 | |
| Muzio (129) | Continuous IR on-line | 2 | |
| CERL (156) | PVC bag (Condenser) | 32 | 39 |
| CERL (156) | PVC bag (Condenser) | 26 | |
| EKONO (156) | PE-lined bags | 11 | |
| EKONO (156) | PE-lined bags | 3 | |
| EKONO (156) | PE-lined bags (Silica gel) | 27 | |
| SERI (156) | PE-lined bags (Silica gel) | 3-60 | |
| SCS (156) | SS and glass bombs | 100-400 | |
| **TANGENTIALLY-FIRED** | | | |
| Kokkinos (92) | On-line | 2 | |
| Kokkinos (92) | On-line | 1 | |
| Kokkinos (92) | On-line | 1 | |
| Kokkinos (92) | On-line | 1 | |

| | | |
|---|---|---|
| Sloan (167) | GC | 4.5 |

## Table 3-1

(Continued)

| Source (Reference) | Analysis Method (Water Removal) | $N_2O$ Content ppm dry, 3% $O_2$ | Boiler Type |
|---|---|---|---|
| Sloan (167) | NDIR1 | 10 | |
| Montgomery (125) | Continuous | <1 | |
| Montgomery (125) | Continuous | <1 | |
| Montgomery (125) | Grab sample with NaOH added | <2 | |
| Montgomery (125) | Grab sample with NaOH added | <2 | |
| Muzio (129) | Continuous IR on-line | 1 | |
| Muzio (129) | Continuous IR on-line | 1 | |
| Muzio (129) | Continuous IR on-line | <1 | |
| Muzio (129) | Continuous IR on-line | <1 | |
| CERL (156) | PVC bag on-line (Condenser) | 2.4 | |
| CERL (156) | PVC bag on-line (Condenser) | 4.6 | |
| ENEL (156) | On-line (Condenser) | 5.1 | |
| ENEL (156) | On-line (Condenser) | 2.1 | |
| ENEL (156) | On-line (Condenser) | 3.3 | |
| ENEL (156) | On-line (Condenser) | 2.5 | |
| SERI (156) | PE-lined bags (Silica gel) | 3,7 ← 2 data points? | ~~or 3.7~~ |
| SCS (156) | SS and glass bombs | ~~100-400~~ | |
| FERCO (156) | Glass bomb | 130 | |
| EPRI (156) | Glass bomb ($H_2O/SO_2$ removed) | 3 | |
| **LOW $NO_x$ BURNERS** | | | |
| Sloan (167) | NDIR1 | 16 | Wall-fired |

put this column before $N_2O$

|  |  |  |  |
|---|---|---|---|
| Sloan (<u>167</u>) | GC | 2.3 | Corner-fired |

Table 3-1

(Continued)

| Source (Reference) | Analysis Method (Water Removal) | $N_2O$ Content ppm dry, 3% $O_2$ | Boiler Type |
|---|---|---|---|
| **FLUIDIZED BED** | | | |
| EKONO (156) | PE-lined bags (Silica gel) | 96 | Circulating |
| EKONO (156) | PE-lined bags (Silica gel) | 80 | Circulating |
| EKONO (156) | PE-lined bags (Silica gel) | 23 | Bubbling |
| SERI (156) | PE-lined bags (Silica gel) | 137 | Bubbling |
| SERI (156) | PE-lined bags (Silica gel) | 126 | Circulating |
| SERI (156) | PE-lined bags (Silica gel) | 88 | Circulating |
| SERI (156) | PE-lined bags (Silica gel) | 106 | Circulating |
| SERI (156) | PE-lined bags (Silica gel) | 79 | Circulating |
| SERI (156) | PE-lined bags (Silica gel) | 165 | Circulating |
| GES (156) | PE-lined bags (Silica gel) | 49,56 | Circulating |
| S.E. (156) | Tedlar bags (Silica gel) | ~~123-209~~ | Circulating |
| S.E. (156) | Tedlar bags (Silica gel) | ~~233-444~~ | Circulating |
| RWE (156) | SS/glass bomb (Condenser) | 115 | Pressurized |
| RWE (156) | SS/glass bombs (Condenser) | 193 | Pressurized |
| RWE (156) | SS/glass bombs (Condenser) | 168 | Pressurized |
| RWE (156) | SS/glass bombs (Condenser) | 95 | Pressurized |
| RWE (156) | SS/glass bombs (Condenser) | 95 | Pressurized |
| VGB (156) | On-line IR | 100 | |
| **BROWN COAL (FLUIDIZED BED)** | | | |
| RWE (156) | SS/glass bombs (Condenser) | 5-11 | Pressurized |

| | | | |
|---|---|---|---|
| RWE (156) | SS/glass bombs (Condenser) | ~2 | Pressurized |
| RWE (156) | SS/glass bombs (Condenser) | 5-8 | Pressurized |

If <, assume = but ind. in column

## Table 3-1

(Continued)

| Source (Reference) | Analysis Method (Water Removal) | $N_2O$ Content ppm dry, 3% $O_2$ | Boiler Type |
|---|---|---|---|
| **STOKER** | | | |
| EKONO (156) | PE-lined bags (Silica gel) | 20 | |
| **OTHER** | | | |
| Linak (106) | On-line | 4.9 | DF Tunnel |
| Linak (106) | On-line | 2.5 | DF Tunnel |
| Linak (106) | On-line | 4.7 | DF Tunnel |
| Linak (106) | On-line | 2.6 | DF Tunnel |
| EPA (156) | SS bomb (Condenser) | ~~10-220~~ | DF Tunnel |
| EPA (156) | On-line (Condenser) | <10 | DF Tunnel |
| UA/DOE (156) | Glass bomb (Condenser) | <5 | DF Tunnel |
| Linak (106) | On-line | 1.38 | DF Tunnel |
| Linak (106) | On-line | 2.06 | DF Tunnel |
| Linak (106) | On-line | 3.91 | UA furnace |
| Linak (106) | On-line | 4.67 | UA furnace |
| Linak (106) | On-line | 1.4 | UA furnace |
| Linak (106) | On-line | 4.7 | Pre-NSPS, Triple |
| Linak (106) | On-line | <4.3 | Pre-NSPS, Circular |
| Linak (106) | On-line | <1.7 | Pre-NSPS, Tangential |

| | | | |
|---|---|---|---|
| Linak (106) | On-line | 0.8 | Pre-NSPS,<br>Tangential |
| Linak (106) | On-line | <1.7 | Pre-NSPS,<br>Tangential |

Table 3-1

(Continued)

| Source (Reference) | Analysis Method (Water Removal) | $N_2O$ Content ppm dry, 3% $O_2$ | Boiler Type |
|---|---|---|---|
| KEMA (156) | TEF-lined bomb (Condenser) | ~1 | |
| KEMA (156) | TEF-lined bomb (Condenser) | ~1 | |
| KEMA (156) | TEF-lined bomb (Condenser) | ~1 | |
| KEMA (156) | TEF-lined bomb (Condenser) | 10 | |
| KEMA (156) | TEF-lined bomb (Condenser) | 20-25 | |
| KEMA (156) | TEF-lined bomb (Condenser) | 3 | |
| KEMA (156) | TEF-lined bomb (Condenser) | 10 | |
| KEMA (156) | TEF-lined bomb (Condenser) | 10-15 | |
| KEMA (156) | TEF-lined bomb (Condenser) | <1 | |
| KEMA (156) | TEF-lined bomb (Condenser) | <1 | |
| KEMA (156) | TEF-lined bomb (Condenser) | 5-8 | |
| KEMA (156) | TEF-lined bomb (Condenser) | 3-6 | |
| L&C S (156) | SS/glass bomb (Condenser) | 27 | Wet bottom |
| L&C S (156) | SS/glass bomb (Condenser) | 25 | Dry bottom |
| L&C S (156) | SS/glass bomb (Condenser) | 4 | Dry bottom |
| VGB (156) | On-line IR | 30 | Combustor |

## Table 3-2

## NATURAL GAS-FIRED COMBUSTOR $N_2O$ EMISSIONS

| Source (Reference) | Analysis Method (Water Removal) | $N_2O$ Content ppm dry, 3% $O_2$ | Boiler Type |
|---|---|---|---|
| **WALL-FIRED** | | | |
| Kokkinos (92) | On-line | 2 | |
| Kokkinos (92) | On-line | 2 | |
| EKONO (156) | PE-lined bags (Silica gel) | 1 | |
| SERI (156) | PE-lined bags (Silica gel) | 2 | |
| SERI (156) | PE-lined bags (Silica gel) | 2 | |
| **TURBINE** | | | |
| Kokkinos (92) | On-line | 4 | |
| Kokkinos (92) | On-line | 1 | |
| **OTHER** | | | |
| KEMA (156) | TEF-lined bomb (Condenser) | ~1 | |
| KEMA (156) | TEF-lined bomb (Condenser) | ~1 | |
| KEMA (156) | TEF-lined bomb (Condenser) | ~1 | |
| Linak (106) | On-line | <0.26 | NA Boiler |
| Linak (106) | On-line | <0.26 | LNB/PBS |
| Linak (106) | On-line | 0.79 | LNB/PBS |

## Table 3-3

## LIGHT OIL-FIRED COMBUSTOR $N_2O$ EMISSIONS

| Source (Reference) | Analysis Method (Water Removal) | $N_2O$ Content ppm dry, 3% $O_2$ | Boiler Type |
|---|---|---|---|
| **WALL-FIRED** | | | |
| Muzio (129) | Continuous IR | <1 | |
| Muzio (129) | Continuous IR | <1 | |
| Kokkinos (92) | On-line | 1 | |
| Kokkinos (92) | On-line | 1 | |
| **UREA INJECTION** | | | |
| Kokkinos (92) | On-line | 23 | |
| **TANGENTIALLY-FIRED** | | | |
| SERI (156) | PE-lined bags (Silica gel) | 23,38 | |
| **OTHER** | | | |
| Linak (106) | On-line | 0.33 | NA boiler |
| Linak (106) | On-line | <0.29 | LNB/PBS |
| Linak (106) | On-line | 0.32 | LNB/PBS |
| SERI (156) | PE-lined bags (Silica gel) | 17,28 | Top-fired |
| SERI (156) | PE-lined bags (Silica gel) | 40 | Top-fired |
| SERI (156) | PE-lined bags (Silica gel) | 3 | Top-fired |
| SERI (156) | PE-lined bags (Silica gel) | 4 | Top-fired |

## Table 3-4

## HEAVY OIL-FIRED COMBUSTOR $N_2O$ EMISSIONS

| Source (Reference) | Analysis Method (Water Removal) | $N_2O$ Content ppm dry, 3% $O_2$ | Boiler Type |
|---|---|---|---|
| **WALL-FIRED** | | | |
| EKONO (156) | PE-lined bags (Silica gel) | 39 | |
| EKONO (156) | PE-lined bags (Silica gel) | 73 | |
| ENEL (156) | On-line (Condenser) | 1 | |
| ENEL (156) | On-line (Condenser) | 1 | |
| **FLUIDIZED BED** | | | |
| GES (156) | PE-lined bags (Silica gel) | 10,12 | Circulating |
| **OTHER** | | | |
| Linak (106) | On-line | 1.4 | NA Boiler |
| Linak (106) | On-line | 0.30 | LNB/PBS |
| Linak (106) | On-line | 0.78 | LNB/PBS |

## Table 3-5

## PEAT-FIRED COMBUSTOR $N_2O$ EMISSIONS

| Source (Reference) | Analysis Method (Water Removal) | $N_2O$ Content ppm dry, 3% $O_2$ | Boiler Type |
|---|---|---|---|
| **WALL-FIRED** | | | |
| EKONO (156) | PE-lined bags (Silica gel) | 20 | |
| EKONO (156) | PE-lined bags (Silica gel) | 18 | |
| EKONO (156) | PE-lined bags (Silica gel) | 21 | |
| **FLUIDIZED BED** | | | |
| EKONO (156) | PE-lined bags (Silica gel) | 43 | Bubbling |
| SERI (156) | PE-lined bags (Silica gel) | 52 | Circulating |
| GES (156) | PE-lined bags (Silica gel) | 6,10 | Circulating |
| GES (156) | PE-lined bags (Silica gel) | 76 | Circulating |
| **STOKER** | | | |
| EKONO (156) | PE-lined bags (Silica gel) | 17 | |
| EKONO (156) | PE-lined bags (Silica gel) | 6 | |
| SERI (156) | PE-lined bags (Silica gel) | 9-18 | |

Table 3-6

## OTHER FUEL-FIRED COMBUSTOR $N_2O$ EMISSIONS

| Source (Reference) | Analysis Method (Water Removal) | $N_2O$ Content ppm dry, 3% $O_2$ | Boiler Type |
|---|---|---|---|
| **WALL-FIRED** | | | |
| EKONO (156) | PE-lined bags (Silica gel) | 29 | Peat/nat. gas |
| **FLUIDIZED BED** | | | |
| EKONO (156) | PE-lined bags (Silica gel) | 7 | Circulating, Peat/wood |
| EKONO (156) | PE-lined bags (Silica gel) | 16 | Bubbling, Peat/oil |
| GES (156) | PE-lined bags (Silica gel) | 62,69 | Circulating, Coal/peat |
| **STOKER** | | | |
| EKONO (156) | PE-lined bags (Silica gel) | 34 | Coal/wood |
| **OTHER** | | | |
| SERI (156) | PE-lined bags (Silica gel) | 1-2 | Rec. furnace, Black liquor |

## Table 3-7

## BIOMASS-FIRED COMBUSTOR $N_2O$ EMISSIONS

| Source (Reference) | Analysis Method (Water Removal) | $N_2O$ Content ppm dry, 3% $O_2$ | Boiler Type |
|---|---|---|---|
| **FLUIDIZED BED** | | | |
| GES (156) | PE-lined bags (Silica gel) | 14,19 | Wood waste |
| **STOKER** | | | |
| EKONO (156) | PE-lined bags (Silica gel) | 1 | Wood chips |
| EKONO (156) | PE-lined bags (Silica gel) | 1 | Wood chips |
| SERI (156) | PE-lined bags (Silica gel) | 5 | Wood chips |
| SERI (156) | PE-lined bags (Silica gel) | 4 | Wood chips |
| SERI (156) | PE-lined bags (Silica gel) | 3 | Wood chips |
| L&C S (156) | SS/glass bombs (Condenser) | 5 | Mun. refuse |
| **OTHER** | | | |
| SERI (156) | PE-lined bags (Silica gel) | 5,8 | Pre-oven, Wood chips |
| SERI (156) | PE-lined bags (Silica gel) | 5 | Pre-oven, Wood chips |

## Table 3-8

## $N_2O$ EMISSION FACTORS FOR COMBUSTION SOURCES

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| Fuel and Combustor Type | Number of Results Reported | Average $N_2O$ Concentration ppm dry at 3% $O_2$ | Emission Factor lbs $N_2O/10^6$Btu |
|---|---|---|---|
| **COAL-FIRED** | | | |
| Wall-fired | 15 | 29.4 | 0.0375 |
| Tangentially-fired | 24 | 18.2 | 0.0233 |
| Low $NO_x$ | 2 | 9.2 | 0.0117 |
| FBC-Circulating | 10 | 129.7 | 0.1658 |
| FBC-Static | 1 | 23.0 | 0.0294 |
| FBC-Pressurized | 9 | 86.9 | 0.1111 |
| Brown Coal (FBC) | 3 | 5.7 | 0.0072 |
| Stoker | 1 | 20 | 0.0256 |
| **NATURAL GAS** | | | |
| Wall-fired | 5 | 1.8 | 0.0021 |
| Turbine | 2 | 2.5 | 0.0029 |
| **LIGHT OIL** | | | |
| Wall-fired | 4 | 1 | 0.0282 |
| Urea injection | 1 | 23 | 0.1064 |
| Tangentially-fired | 1 | 30.5 | 0.0069 |
| **HEAVY OIL** | | | |
| Wall-fired | 4 | 28.5 | 0.0347 |

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OK

| | | | |
|---|---|---|---|
| FBC-Circulating | 1 | 11 | 0.1340 |

## Table 3-8

(Continued)

| Fuel and Combustor Type | Number of Results Reported | Average $N_2O$ Concentration ppm dry at 3% $O_2$ | | Emission Factor lbs $N_2O/10^6$Btu |
|---|---|---|---|---|
| **PEAT** | | | | |
| Wall-fired | 3 | 19.7 | 845.5 | 0.0233 |
| FBC-Static | 1 | 43 | [illegible] | 0.0510 |
| FBC-Circulating | 3 | 45.3 | [illegible] | 0.0144 |
| **WOOD** | | | | |
| FBC-Circulating | 1 | 16.5 | 841.8 | 0.0196 |
| Stoker | 6 | 3.2 | [illegible] | 0.00375 |

~~$\frac{.00375 \text{ lb } N_2O}{10^6 \text{ BTU}} \times \frac{1050 \text{ BTU}}{\text{SCF}} = \frac{3.94 \text{ lb}}{10^6 \text{ SCF}}$~~

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