

# Nitrous Oxide Emissions and the Anthropogenic Nitrogen in Wastewater and Solid Waste

Philip K. Barton<sup>1</sup> and James W. Atwater<sup>2</sup>

**Abstract:** In the 20th century, human interference in the nitrogen cycle has caused a doubling of the global nitrogen fixation rate (an element critical in the proteins of all organisms), thereby intensifying global nitrous oxide (N<sub>2</sub>O) production during microbial nitrification and denitrification. Nitrous oxide is a powerful greenhouse gas, important in climate change, and as well, is a stratospheric ozone-depleting substance. It is likely that much of the Earth's population now relies on anthropogenic nitrogen in its food supplies, resulting in anthropogenic nitrogen contained in wastes requiring management. Food production is considered as a source of global nitrous oxide emissions; however, the nitrogen in wastewater and solid wastes may be a significant fate of much anthropogenic nitrogen. This factor has largely escaped in-depth, critical analysis from the perspective of nitrous oxide emissions. This paper introduces nitrogen cycling and nitrous oxide production and reviews the research currently available on N<sub>2</sub>O emissions from wastewater treatment operations, landfilling, composting, and incineration; demonstrating that each process can emit large amounts of this important gas. This is followed by a discussion of the limited research. The relative importance of N<sub>2</sub>O in waste management is also estimated, indicating that wastewater treatment may be the most important operation for managing anthropogenic nitrogen in wastes.

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## Introduction

The human population has grown at an unprecedented rate this century and this has resulted in many localized environmental impacts. There is now believed to be potential for the population to start interfering with the atmosphere, with global-scale environmental impacts being the result. In this decade, human impacts on the carbon cycle, primarily from fossil fuel combustion and deforestation, have received much public and governmental attention because of the implications for enhancing the natural greenhouse effect. However, less attention has been given to human impacts on the global nitrogen cycle, impacts which are quantitatively greater than the impacts on the carbon cycle. Fortunately, several members of the scientific community have recently published reviews raising concerns about the human-induced impacts on global nitrogen cycling (Vitousek and Matson 1993; Ayres et al. 1994; Isermann 1994; Kinzig and Socolow 1994; Galloway et al. 1995; Galloway 1998). Analogous to the increase in atmospheric carbon dioxide concentrations being labeled, "global warming," the impact of human-induced interference of the global nitrogen has been called "global fertilization." Unfortunately,

interference on global nitrogen creates not only global fertilization, but also contributes to global warming and stratospheric ozone depletion.

This paper will examine the importance of the nitrogen contained in solid waste and wastewater on human-induced global environmental impacts. This nitrogen is the 25% of agriculturally applied nitrogen that leaves the farm as food. The paper is written primarily for the waste management community in order to stimulate awareness of the inter-relationships that exist and to call for research in this field. The paper will briefly review the global biogeochemical cycling of nitrogen and the implications for waste management. This paper will be specifically concerned with the future implications for wastewater treatment, landfilling, composting, and waste incineration.

## Background

Nitrogen is a critical element for life. It is an essential component of amino acids, and amino acids are the building blocks of protein. It is estimated that 16% of protein, or 0.16 kg of N per kg of protein, is nitrogen (IPCC 1997). The cycling of global nitrogen throughout terrestrial and aquatic ecosystems and the atmosphere is critical for this building block.

## Global Nitrogen Cycle

The Earth's atmosphere is 78% by volume elemental nitrogen N<sub>2</sub> gas and is therefore an immense reservoir of N<sub>2</sub>. It is estimated that  $5 \times 10^{15}$  metric tonnes of nitrogen are contained in the atmosphere, ocean, terrestrial and marine biota, soil organic matter, and sedimentary rocks and though N is abundant, the vast majority of it is in a form which can be used by only a few living organisms (Mackenzie 1998). It is estimated that <0.02% (or  $\sim 1 \times 10^{12}$  tonnes) of global N is actually accessible to living

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organisms as most is either diatomic nitrogen ( $N_2$  gas) in the atmosphere ( $\sim 78\%$ ) or tied up in sedimentary rocks ( $\sim 20\%$ ) (ibid). To be available for utilization by living organisms, it must be in the form of reactive or fixed nitrogen [defined as “N bonded to C, O, or H (e.g.,  $NO_y$ ,  $NH_x$ , organic N)”] (Galloway 1998). Only a few species of aquatic and terrestrial bacteria and blue-green algae can fix the nearly inert  $N_2$  molecule into ammonium ( $NH_4^+$ ) for utilization (Smil 1999). This conversion is called biological N fixation and it has been estimated that in preindustrial times  $(90-130) \times 10^6$  metric tonnes of N was annually fixed by this natural process (Galloway 1995). Lightning contributes an estimated additional  $(3-5) \times 10^6$  metric tonnes of N annually.

This reactive or fixed nitrogen moves through the terrestrial N cycle by the death of plants and microorganisms, *mineralization*—the breakdown of amino acids, *assimilation*—uptake by plants and by *immobilization*—uptake by microorganisms (Kinzig and Socolow 1994). Assuming an annual fixation rate of 130 million tonnes, this is minuscule in relation to the pool of total nitrogen (1 part in 38 million) or to the pool of reactive nitrogen (1 part in 7800); however, it is upon this fixation rate which organisms depend. Since plant and animal life can only utilize reactive nitrogen as nutrients, and not diatomic nitrogen, “all life ultimately depends on nitrogen fixation” (Ayres et al. 1994).

For nitrogen to remain in steady-state conditions, there must also be the return of reactive N to the  $N_2$  reservoir (though this reactive N may transfer many times due to oxidations and reductions before returning to  $N_2$ ). The majority of this is accomplished by the microbial processes, *nitrification* and *denitrification*. There is also *pyrodenitrification*, which occurs during forest fires (Ayres et al. 1994). In nitrification, ammonium ( $NH_4^+$ ) is oxidized to nitrate ( $NO_3^-$ ) by mainly autotrophic bacteria under aerobic conditions in order to derive energy. In denitrification, nitrate is reduced to  $N_2$  by mainly facultative heterotrophic bacteria under anoxic (oxygen restricted) conditions. These bacteria use the nitrate as a terminal electron acceptor so that they can concurrently derive energy from the breakdown of organic carbon (Beauchamp 1997). During pyrodenitrification, “two nitrogen atoms that were in separate molecules in plants or soil bind to one another to make  $N_2$ , as the result of a sequence of high-temperature reactions” (Ayres et al. 1994). By these three processes, equal amounts of N (the approximately 93–135 Tg N fixed annually) is returned to the atmosphere as  $N_2$  gas.

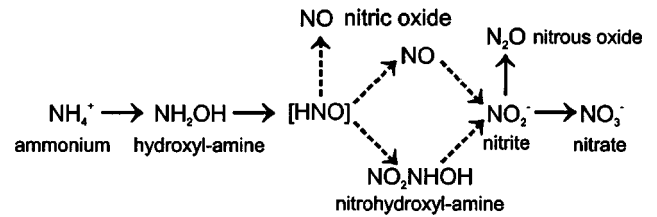
### Global Nitrous Oxide Production

Nitrous oxide ( $N_2O$ ) is a stable gas which leaks from microbial cells during nitrification and denitrification (Firestone and Davidson 1989). Though the predominant result of nitrification is nitrate ( $NO_3^-$ ), and of denitrification is  $N_2$ , a portion of the N can be emitted as nitrous oxide.

Though some parts of the nitrification and denitrification pathways are not completely understood, primarily the pathways encompassing nitric oxide (NO), the generally accepted metabolic pathways are shown below. (Hooper 1984, Firestone and Davidson 1989). Recent work suggests that nitric oxide is an obligate intermediate (Krause and Neelson 1997). See Fig. 1.

The small leakage takes on importance because  $N_2O$  is a strong greenhouse gas (GHG). It is estimated to be 310 times more effective than carbon dioxide as a greenhouse gas (based on a 100 year timeframe as recommended by the Intergovernmental Panel on Climate Change [(IPCC 1995)]). In addition,  $N_2O$  catalytically destroys stratospheric ozone and is largely responsible

### NITRIFICATION (Hooper 1984; Firestone and Davidson 1989):



### DENITRIFICATION (Firestone and Davidson 1989):

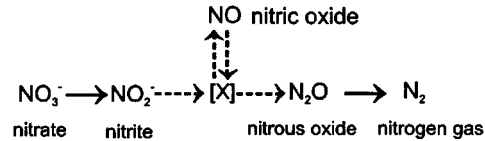


Fig. 1. Nitrification and denitrification pathway

for naturally regulating stratospheric  $O_3$  concentrations (Abbatt and Molina 1993).

The “hole-in-the-pipe” or “process-pipe” conceptualization by Firestone and Davidson allows one to visualize  $N_2O$  production in soils. These researchers consider that  $N_2O$  production is a factor of (1) the amount of nitrogen cycling between the soil-plant-microbial system, and (2) the ratios of the  $N_2O/NO_3^-$ , and  $N_2O/N_2$  products of nitrification and denitrification. These two factors, the overall movement of nitrogen through the *pipe*, and the amount of *leakage* (the size of the holes in the pipe), control the emission of  $N_2O$ .

Agricultural research has found that anywhere from 0.001 to 6.8% of the nitrogen applied to fields is emitted as  $N_2O$  (Mosier et al. 1996) and the ratio of  $N_2O/NO_3^-$  produced as a result of nitrification has been reported as high as 20% (Martikainen 1985), but is generally below 1% (Firestone and Davidson 1989). Though both processes have been demonstrated to result in  $N_2O$  leakage, denitrification is considered to be the major source of  $N_2O$  in soils (Sahrawat and Keeney 1986).

The production of  $N_2O$  during nitrification in soils has been demonstrated to result from

“A reductive process in which the organisms use  $NO_2^-$  as an electron acceptor, especially when  $O_2$  is limiting. This mechanism not only allows the organisms to conserve limited  $O_2$  for the oxidation of  $NH_4^+$  (from which they gain energy for growth and regeneration), but also avoids the potential for accumulation of toxic levels of  $NO_2^-$ ” (Poth and Focht 1985); (Hutchinson and Davidson 1993).

The nitrous oxide leakage occurring during denitrification is considered to result when

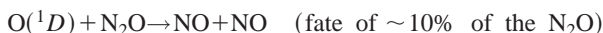
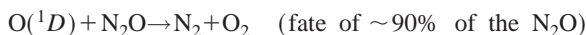
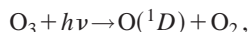
“The availability of oxidant (N oxide) greatly exceeds the availability of reductant (most commonly organic carbon), then the oxidant may be incompletely utilized, i.e.,  $N_2O$  will be produced” (Firestone and Davidson 1989). “Conversely, when the overall rate of denitrification is limited by the supply of oxidant, most of the N oxide is converted to  $N_2$ ” (Hutchinson and Davidson 1993).

The estimate of the IPCC (1995) is that  $9 \times 10^6$  tonnes of  $N_2O$ -N is naturally emitted to the atmosphere by soil processes, forest and brush fires, and the oceans. As there is no chemical loss in the troposphere, this  $9 \times 10^6$  tonnes of  $N_2O$ -N slowly rises to the stratosphere where it is destroyed. The destruction of  $N_2O$  occurs at an altitude above 30 km (in the stratosphere) and returns this nitrogen to the  $N_2$  reservoir. It takes, on average, 120 years

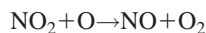
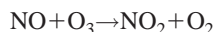
**Table 1.** Global Reactive Nitrogen Sources

	Global nitrogen fixation (Tg N/year)
Natural biological N fixation	90–130
Lightning	3–5
Natural source subtotal	93–135
Synthetic fertilizers	80–90
Human-induced biological N fixation	27–44
Fossil fuel combustion	30
Anthropogenic source subtotal	137–164
Total sources	230–299

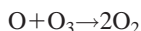
for  $N_2O$  to reach this altitude and to be destroyed (IPCC 1995). The  $N_2O$  is destroyed predominantly by photodissociation into  $N_2$  and O atoms. However, approximately 10% of the  $N_2O$  reacts with electronically excited oxygen atoms (formed by the photolysis of ozone) to form NO. This is demonstrated in the following reactions (Abbatt and Molina 1993):



The production of NO is important in stratospheric ozone chemistry, since NO catalytically destroys ozone



Net



### Anthropogenic Interference of the Global Nitrogen Cycle

The definition of *anthropogenic* is “involving the input of man on nature: induced or altered by the presence and activity of man.” (Websters 1967) The definition of *interference* is “the act of *interfering*” and the definition of *interfering* is “to come in collision: to be in opposition” (ibid). By putting all these definitions together, one would hope to come up with the meaning of *anthropogenic interference*, and it could be expected to be “human-induced collisions with natural processes.” While this definition may seem logical it does not adequately explain the current state of anthropogenic interference with the global nitrogen cycle.

Humans have approximately doubled the global rate of N fixation, from an estimated pre-industrial rate of 93–135 TgN/yr to an estimated 243–295 TgN/yr fixed currently (Galloway 1998). Thus, anthropogenic interference is not a collision with natural processes, but in fact, is the supplementing or boosting of natural processes (but is interference no less). The increased fixation of elemental  $N_2$  gas has been a direct result of nitrogenous fertilizer production, human induced increases in the cultivation of leguminous crops (which host a symbiotic relationship with biological N-fixing bacteria) and by the combustion of fossil fuels. Estimates adapted from Galloway (1998) of this anthropogenic interference are provided in Table 1.

Fertilizer production by the Haber-Bosch process, fixes  $N_2$  to ammonia ( $NH_3$ ) thereby reproducing at high temperature and pressure what bacteria can accomplish with enzymes at ambient

temperature and pressure (Kinzig and Socolow 1994). Human-induced biological N fixation occurs by the increased cultivation of leguminous crops such as soybean and alfalfa, which symbiotically host nitrogen fixing bacteria in their root nodules. The last source of reactive N occurs accidentally as a side reaction of the combustion of fossil fuels. Whereas nitrogen fixation from fertilizer production and the cultivation of leguminous crops was intended, the reactive N produced as a result of combustion is not intended. The combustion of fossil fuels causes nitrogen that was originally in air as  $N_2$  gas or nitrogen that was originally sequestered in the fuel as organic N to be oxidized to nitric oxide (NO) or nitrogen dioxide ( $NO_2$ ).

Previously mentioned was the fact that nitrogen is an essential element of life as it is a critical component of protein. Clearly, the recent doubling of the rate of global N fixation has the potential for large and long-range environmental impacts. The term “global fertilization” has been labeled to identify the potential for the stimulation of unintended growth in natural terrestrial and aquatic ecosystems. A long list of the negative impacts of excess nitrogen has been compiled by Cowling et al. (1998). A small part of this is

- “Acidification of soils, lakes, streams and ground waters;
- Eutrophication of freshwater lakes and ecosystems;
- Blooms of toxic algae and decreases in “swimability” of water bodies;
- Nitrogen saturation of forest soils; and
- Biodiversity impacts on ecosystems”

### Anthropogenic Interference of the Global Nitrous Oxide Cycle

As nitrous oxide production is a side function of the nitrogen cycling, it is reasonable to suggest that if the global N fixation rate is effectively doubled (thereby doubling the amount of nitrogen which eventually requires denitrification) then  $N_2O$  emissions would increase? This is indeed what is currently happening. As a direct result of the enhancing of the global nitrogen cycle there has been a consequent enhancing of the global nitrous oxide cycle (Vitousek and Matson 1993; Ayres et al. 1994; Kinzig and Socolow 1994; Bakken and Bleken 1998).

The atmospheric  $N_2O$  concentration has increased from a pre-industrial level of 275 ppb to a level of 311 ppb in 1992, an increase of 13% (IPCC 1995). Preliminary data suggests that global averages are about 316 ppb (NOAA 2000). This is estimated to contribute 6% of the human-induced increase in radiative forcing of the atmosphere (Erisman et al. 1998). In addition, this concentration continues to increase by about 0.8 ppb annually (IPCC 1995).

Also, the fate of approximately 10% of  $N_2O$  molecules is to become nitric oxide (NO) and catalytically destroy stratospheric ozone. As the result of increased use of nitrogen fertilizer by agriculture, it has been estimated by Crutzen and Ehhalt (1977) that the subsequent  $N_2O$  emissions could result in a 10% decrease in the ozone layer by the end of the 21st century. However, the current anthropogenically induced ozone losses are not believed to be primarily due to nitrous oxide but to long-lived industrial chlorine and bromine-containing compounds, such as chlorofluorocarbons (Kinzig and Socolow 1994).

The most recent budget, as of 1998, of global nitrous oxide is provided in Table 2 (adapted from Mosier et al. 1998).

Agricultural activities dominate the emissions of anthropogenic nitrous oxide with industrial sources (primarily fossil fuel combustion) providing the remainder of emissions.

**Table 2.** Global N<sub>2</sub>O Budget

	Global sources and sinks of N <sub>2</sub> O (Tg N <sub>2</sub> O-N/year)
Identified natural sources	9.0 (4.3–14.7)
Identified anthropogenic sources	7.2 (2.1–19.7)
Total identified sources	16.2 (6.4–34.4)
Unaccounted sources	0
Total sinks (photolysis)	12.3 (9.0–16.0)
Atmospheric increase	3.9 (3.1–4.7)

It is only since 1998 that the budget of N<sub>2</sub>O sources and sinks has been successfully balanced. Previous budgets were not balanced because sinks exceeded sources with the missing emissions labeled as “unaccounted sources.” In the newest IPCC methodology, there are sources of N<sub>2</sub>O which were previously not included (IPCC 1997). In this new methodology, “three sources of N<sub>2</sub>O are distinguished: (1) direct emissions from agricultural soils, (2) emissions from animal production systems, and (3) N<sub>2</sub>O emissions indirectly induced by agricultural activities” (IPCC 1997; Mosier et al. 1998; Kroeze et al. 1999). It is the N<sub>2</sub>O emissions indirectly resulting from agriculture with which this paper is concerned.

Indirect emissions of N<sub>2</sub>O, from a subsequent waste management perspective, are caused by reactive N exported from farms in the form of food products being eventually nitrified and denitrified—thereby, resulting in N<sub>2</sub>O leakages. These sources of N<sub>2</sub>O are considered indirect because they do not actually occur on farms but are nevertheless, agricultural in origin. For instance, the nitrification and denitrification of reactive N used in food production (and the associated N<sub>2</sub>O leakage) may occur nowhere near agricultural fields; to illustrate, the nitrification and denitrification (and N<sub>2</sub>O emissions) may occur at wastewater treatment plants. As a result, N<sub>2</sub>O emissions from wastewater treatment can be considered indirectly agricultural in origin, in fact, any N<sub>2</sub>O emissions from reactive N downstream of food production are indirectly agricultural N<sub>2</sub>O emissions. This will be further discussed in the “Implications for Waste Management” section.

To give the reader a feel for the N<sub>2</sub>O losses currently being debated, emission factors estimated by the IPCC (1997) are provided below

- Synthetic fertilizers 0.0125 (0.0025–0.0225) kg N<sub>2</sub>O-N/kg N input;
- Nitrogen deposition 0.01 (0.002–0.02) kg N<sub>2</sub>O-N/kg NH<sub>3</sub>-N and NO<sub>x</sub>-N emitted; and
- Leaching and runoff 0.025 (0.002–0.12) kg N<sub>2</sub>O-N/kg N leaching/runoff.

The first is the direct losses estimated to occur as a result of land application of nitrogenous fertilizer. The second is an indirect loss to be expected when reactive nitrogen (NH<sub>3</sub> or NO<sub>x</sub>) is emitted to the atmosphere and is subsequently deposited on land downwind. The third, also indirect, is from the leaching and/or runoff of reactive nitrogen from agricultural fields.

As explained, there is substantial anthropogenic interference in nitrogen fixation and nitrous oxide emissions, yet it will be discussed in the next section that we may not even be experiencing the full effect of our actions.

### Transient Versus Steady-State Conditions

It is possible that the full anthropogenic impact on the global nitrogen cycle is not yet reflected in current N<sub>2</sub>O emissions. Even

if all human-induced nitrogen fixation was to immediately cease, anthropogenic emissions of N<sub>2</sub>O may still continue to occur for many years. This is because we are more likely experiencing a transient period of reactive N accumulation; there is more N being annually fixed by natural and human-induced means than is being denitrified. It is estimated that the forested ecosystems are accumulating N, the nitrate concentrations of ground water are increasing, and the sedimentation rates of organic matter have been accelerated (Galloway 1998). If part of the anthropogenic reactive N fixed annually is currently accumulating in a sort of *nitrogen storage*, we are not yet experiencing the full anthropogenic N<sub>2</sub>O emissions which must follow—for steady-state conditions to once again be attained, this reactive N will eventually have to be nitrified and denitrified. This important concept was well explained by Bleken and Bakken (1996):

“The likely outcome of the increased input [of reactive nitrogen] is a transient period of net accumulation of biologically available nitrogen (primarily as organic nitrogen). It will be transient, because removal through denitrification (and possibly by “fossilization” in sediments) *sooner or later will balance the level of input and therefore reach steady-state conditions*” (emphasis added).

When steady-state conditions are eventually reached, the full anthropogenic emissions of N<sub>2</sub>O will be realized. This could be higher than the current estimated emissions as modeling by Bakken and Bleken (1998) has suggested that future long-term emissions are routinely underestimated. Readers will remember that Galloway (1998) had estimated the annual nitrogen fixation as being between 93 and 135 × 10<sup>6</sup> tonnes and that the IPCC (1995) estimates 9.0 × 10<sup>6</sup> tonnes of N<sub>2</sub>O-N is emitted annually as part of the natural (preindustrial) global nitrous oxide cycle. If we assume that 110 × 10<sup>6</sup> tonnes of N is naturally fixed annually (and 110 × 10<sup>6</sup> tonnes of N has to be annually denitrified to remain at steady-state conditions) the 9 × 10<sup>6</sup> tonnes of N<sub>2</sub>O-N represents 8.2% of the annual fixed and subsequently denitrified natural nitrogen. Therefore, when our planet reached a preindustrial steady state, 8.2% of reactive N upon nitrification and/or denitrification was diverted to the global N<sub>2</sub>O cycle. Yet the current estimate of anthropogenic N<sub>2</sub>O emission is 7.2 × 10<sup>6</sup> tonnes N<sub>2</sub>O-N (with a range of (2.1–19.7) × 10<sup>6</sup> tonnes) from an anthropogenic N fixation (150–160) × 10<sup>6</sup> tonnes N (estimated by Galloway 1998). The percentage of 7.2 into 155 is only 4.6%, though this is undoubtedly crude, could it be indicating that we have yet to experience the full N<sub>2</sub>O emissions to result from our current level of nitrogen fixation?

### Implications for Waste-Management

The existence of anthropogenic nitrogen in our food supply has important implications for the management of the nitrogen in food-derived waste. If the nitrogen in food is anthropogenic in origin, then any N<sub>2</sub>O emissions resulting from either wastewater treatment, composting, landfilling, or incineration of these wastes is anthropogenic. Thus, any direct or indirect N<sub>2</sub>O emissions resulting from food waste management is anthropogenic interference of the global N<sub>2</sub>O.

It is quite likely that the majority of the nitrogen in food products consumed by developed countries is anthropogenic in origin. This could be particularly true where the nitrogen content of the soil is not being drawn down. That is, it resulted from either synthetic fertilizers or human-induced biological N fixation by legume cultivation. In a nitrogen study by Bleken and Bakken

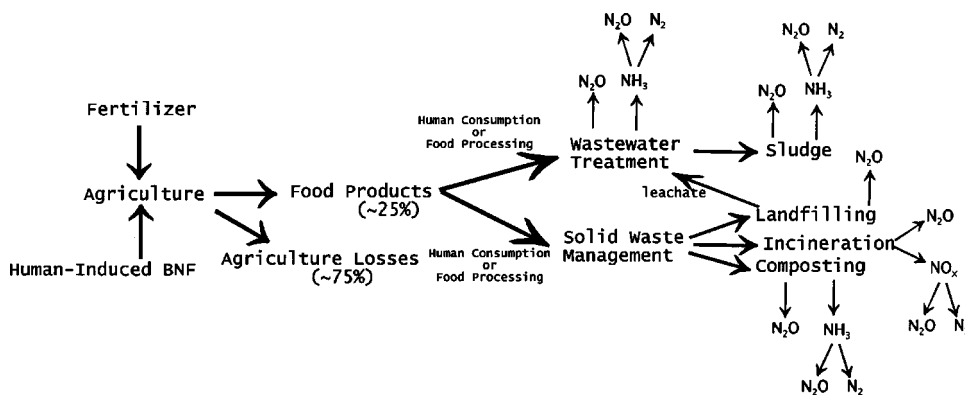


Fig. 2. Conceptualization of N<sub>2</sub>O emissions from waste management

(1997), the only identified sources of natural N consumed by Norwegian society represented less than 10% of inputs. These were wild seafood and wild animals such as moose and lamb, which were raised on natural, noncultivated pastures in mountains. For simplicity, it will be assumed for the remainder of this paper that all of the reactive N in food products is human induced. As a result, any N<sub>2</sub>O emissions from the waste management of this material, either after human consumption or food processing, can be considered anthropogenic greenhouse gas emissions.

Recently, several researchers have estimated the nitrogen efficiency of their respective countries agricultural industry (efficiency of N in food products versus total N input required). The results of these overall N efficiency studies vary a great deal, 10% for Norway (Bleken and Bakken 1997) and 25% for Germany (Isermann and Isermann 1998), primarily because of the methodology used. In Germany, a portion of the N input is from imported animal feed produced in other countries, and this study does not include the N losses from plant production in the exporting countries. However, the Norwegian study was corrected for any imported animal feed to include all the N losses. In addition, the German study considered food before the N losses from food processing while the Norwegian study considered food after processing. Overall, Isermann and Isermann (1998) has estimated that the N efficiency of European Union agriculture as only being between 20 and 30%. For discussion purposes, let us assume that the N efficiency of agriculture is about 25% in developed countries prior to food processing.

With the assumptions that all nitrogen in food is anthropogenic and that 25% of nitrogen inputs to agriculture becomes food products carried forward, what is to become of this 25% nitrogen in food? While there has been hundreds of research papers published internationally in over ten years of considerable effort investigating the 75% N lost from agriculture, resulting in the N<sub>2</sub>O emissions from fertilizer and manure applications, there has not been a comparable level of research into the 25% N leaving fields as food products. As this 25% N in food products eventually winds up as waste or wastewater after human consumption or food processing, there should be a comparative effort. A survey of the research literature has found only 26 papers and one patent on the N<sub>2</sub>O emissions from wastewater treatment, ten papers on N<sub>2</sub>O emissions from waste incineration, eight papers on the N<sub>2</sub>O emissions from composting, and only two papers on the N<sub>2</sub>O emissions from waste disposed in landfills. (It needs to be noted that the authors have not been successful in acquiring all of these papers.) While on a mass basis, N<sub>2</sub>O emissions are an irrelevant

component of waste management, they are of great importance when investigating greenhouse gases.

These papers actually generate many more questions than they answer. For all of the papers, only the immediate emissions of N<sub>2</sub>O were measured during the investigated process. There was no attempt to quantify any *future emissions* resulting from ammonia, nitric oxide, or nitrate losses during the process or from the future decomposition of the organic matter. For example, gaseous emissions of NH<sub>3</sub> are common during wastewater treatment or composting, but losses of this reactive nitrogen are not considered with respect to subsequent N<sub>2</sub>O emissions.

Ammonia has an atmospheric lifetime of only a few hours to a few days and is mainly removed by wet and dry deposition, however a portion is oxidized to NO<sub>x</sub>, with a third fraction reacting with nitrates or sulphates to form ammonium-containing aerosols, (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and NH<sub>4</sub>NO<sub>3</sub>. These aerosols are later removed by rainfall or dry fallout (Matthews 1994). The NO<sub>x</sub> is short lived with an atmospheric lifetime between 1 and 10 days (Olivier et al. 1998). Nitrogen oxides are removed from the atmosphere by conversion to nitric acid (HNO<sub>3</sub>) which is followed by wet or dry deposition (Logan 1983; Olivier et al. 1998). In addition, nitrogen oxides are suspected to indirectly contribute to global warming—they deplete the upper atmospheric concentration of the OH radical; a radical which limits the atmospheric lifetime of CH<sub>4</sub>. With OH depleted, the potential exists for methane, a greenhouse gas estimated to be 21 times more powerful than CO<sub>2</sub>, to have a longer atmospheric lifetime than it otherwise would be able to (Mackenzie 1995).

Once these nitrogen compounds (NH<sub>3</sub>, NH<sub>4</sub><sup>+</sup>, (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, NH<sub>4</sub>NO<sub>3</sub>, HNO<sub>3</sub>) are returned to land or water surfaces by wet or dry deposition, this reactive nitrogen is again available for nitrification/denitrification and the associated N<sub>2</sub>O emissions can result. As a result, losses of reduced or oxidized nitrogen compounds only temporally and spatially alter the nitrous oxide emissions which result, but does nothing to prevent these emissions (Mosier et al. 1996; Ferm 1998).

Another example of a *future N<sub>2</sub>O emission* originates from the organic nitrogen contained in wastewater sludge of finished compost. As this organic nitrogen will eventually undergo decomposition, the released ammonium (NH<sub>4</sub><sup>+</sup>) is now available to undergo nitrification and denitrification. A simplified conceptualization of N<sub>2</sub>O emissions during waste management operations and of potential future emissions is demonstrated in Fig. 2.

Research needs to begin to fully quantify these emissions. There needs to be full nitrogen balances of all the operations of waste management as a system, i.e., all the flows of reactive N into the system and all of the flows of reactive N, N<sub>2</sub>O, and inert N<sub>2</sub> out of the system need to be critically analyzed. Specifically, any flows of reactive N out of a system need to be considered as potential sources of N<sub>2</sub>O as the reactive N will eventually undergo nitrification and denitrification. It is this type of research which is necessary for the waste management community to comprehensively address N<sub>2</sub>O emissions.

From the perspective of the operation of wastewater treatment, landfilling, incineration or composting, there are really only two options to mitigate N<sub>2</sub>O emissions (1) maximizing the conversion of reactive N to N<sub>2</sub> while concurrently minimizing the leakage of N<sub>2</sub>O gas (herein defined as “clean denitrification”), or (2) recycling of reactive N back to agriculture to reduce the requirement for new reactive N. The first option may be performed by ensuring the O<sub>2</sub> and carbon levels are optimal for N<sub>2</sub> production in the case of microbial nitrification and denitrification during wastewater treatment or composting and controlling the temperature range during waste combustion. The recycling of N back to agriculture could conceivably prevent N<sub>2</sub>O emissions by preventing the fixation of N for fertilizers which would otherwise be necessary. This is well stated by De Kimpe and MacDonald (1998) “Ultimately, the recycling of nutrients from all sources, e.g., livestock manures, municipal organic waste compost, and sewage sludge, back into agricultural systems will reduce reliance on nonrenewable nitrogen sources and contribute to sustainability.”

Recycling nitrogen would also prevent the carbon-based greenhouse gas emissions associated with the manufacture of nitrogenous fertilizers from fossil fuels estimated by Environment Canada to be 1.6 kg of CO<sub>2</sub> per kg of NH<sub>3</sub> (Jaques et al. 1997). While this may appear logical, the tremendous transportation demand (and fossil fuel combustion) of moving organic composts and wastes to do this may prevent any greenhouse gas benefit from actually being realized. Until such time when the current carbon-intensive transportation system is replaced, this option may not be attractive. As a result, the “clean” destruction of reactive nitrogen to N<sub>2</sub> gas is likely the best option at present (though the life-cycle greenhouse gas emissions of this recycling option requires investigation). The following sections will discuss the current state of knowledge of N<sub>2</sub>O emissions and discuss the implications of “clean” denitrification-recycling back to agriculture is left for future work.

To reiterate, N<sub>2</sub>O emissions in waste management from the reactive nitrogen in food waste is considered *indirectly* agricultural in origin by the IPCC. This is because the nitrogen was originally fixed for food production. The waste management community needs to begin taking responsibility for these anthropogenic N<sub>2</sub>O emissions. During waste management, there can be *immediate* or *future* emissions of N<sub>2</sub>O. Immediate emissions occur during the operation in question, be it wastewater treatment, composting, landfilling, or incineration. Future emissions occur as a result of the future nitrification and denitrification of reactive N, which exits the operation, usually in the form of ammonia or nitrogen oxides emissions or organic nitrogen in compost or sludge. While waste managers are responsible for the release of reactive nitrogen, once the nitrogen is released, they have no way of controlling the conversion rates to N<sub>2</sub>O.

### Wastewater Treatment

A survey of available research conducted on the N<sub>2</sub>O emissions from wastewater treatment has found 26 papers. Interestingly, the

studies of actual wastewater treatment plants observed very low N<sub>2</sub>O emissions, but laboratory experiments generally reported much higher N<sub>2</sub>O emissions.

Studies using laboratory-scale reactors have demonstrated high N<sub>2</sub>O losses, but have also observed a profound ability to affect these emissions; therefore the possibility exists for mitigation of a large portion of these emissions. The conversion of ammonium-nitrogen (NH<sub>4</sub>-N) to N<sub>2</sub>O during nitrification has been demonstrated by Zheng et al. (1994) to be between 2.3 and 7.0% at dissolved oxygen (DO) concentrations between 0.1 and 6.8 mg/L. They also found N<sub>2</sub>O conversions as high as 16% and as low as 2.3% at solids retention times (SRT) of 3 days and 10 days, respectively. It was concluded that high N<sub>2</sub>O production resulted when incomplete nitrification occurred. Thus, preventing incomplete nitrification by maintaining DO levels greater than 0.5 mg/L and having SRTs of greater than five days would greatly reduce N<sub>2</sub>O emissions. By also using reactors, Hanaki et al. (1992) observed that as much as 8% of influent nitrate nitrogen (NO<sub>3</sub>-N) was transformed to N<sub>2</sub>O during denitrification, though several experiments demonstrated very little N<sub>2</sub>O. The high conversion to N<sub>2</sub>O was observed at a chemical oxygen demand (COD) to NO<sub>3</sub>-N ratio of 3.5, a SRT of 0.5 days and a pH of 6.5. These researchers concluded that “N<sub>2</sub>O production can be avoided by achieving complete denitrification [by maintaining] high COD/NO<sub>3</sub>-N in wastewater, long SRTs and neutral to alkaline pH conditions.” Work by Hong et al. (1993) also agrees with this; they concluded that “the lower the ratio of COD/NO<sub>3</sub>-N, the higher the percentage of N<sub>2</sub>O in the produced gas was” and that short hydraulic retention times resulted in higher N<sub>2</sub>O production.

Several papers have reported extremely high N<sub>2</sub>O conversion rates in the laboratory. Osada et al. (1995) reported a 35% N<sub>2</sub>O-N conversion in a fill and draw activated sludge process while treating swine wastewater under continuous aeration but only a 0.7% N<sub>2</sub>O-N conversion during intermittent aeration. Experiments on a sequencing-batch reactor (SBR) found that as much as 40% of the removed nitrogen was emitted as N<sub>2</sub>O; most of which occurred during the low DO period in aeration (Okayasu et al. 1997). During the aerobic treatment of swine slurry, nitrous oxide emissions could represent up to 30% of the total nitrogen content of the slurry (Beline et al. 1999). Research by Spector (1998a), observed that N<sub>2</sub>O accumulated to a maximum and was subsequently reduced to N<sub>2</sub> gas, when reducing nitrate with methanol in a closed reactor. At the time of maximum accumulation ( $t_{max}$ ), 50 to 80% of the reduced nitrate was in the form of N<sub>2</sub>O. This scientist noted that if the same experiment had been performed in an open reactor, “most or all the N<sub>2</sub>O retained at  $t_{max}$  would have been discharged to the atmosphere.”

Research investigating the mechanisms of N<sub>2</sub>O formation have also developed important results recently. Since nitrous oxide is an intermediate of denitrification and it is reduced to N<sub>2</sub> gas by the enzyme nitrous oxide reductase, inhibition of this enzyme can cause N<sub>2</sub>O to become a significant end product of denitrification. Work by Thörn and Sörensson (1996) at a pilot plant, observed that pH had a considerable influence on the N<sub>2</sub>O production of the denitrification basin. Nitrous oxide production was at its maximum at a pH of 5.5 and undetectable above a pH of 6.5. Research published in 1998 by Schönharting et al. (1998) has a likely answer for this observation—strong pH-dependent inhibition of N<sub>2</sub>O reduction by hydrogen sulfide (H<sub>2</sub>S). They found that it is the free H<sub>2</sub>S and not the total sulfur (total S=H<sub>2</sub>S+HS<sup>-</sup>+S<sup>2-</sup>) which can strongly inhibit the enzyme nitrous oxide reductase; thus, the enzyme inhibition is a function of the pH-dependent dominant species of sulfur present. It was observed that even

H<sub>2</sub>S-S concentrations as low as 0.32 mg/L can lead to a strong inhibition of N<sub>2</sub>O reduction, therefore resulting in a high release of N<sub>2</sub>O from wastewater treatment plants. Or this pH/N<sub>2</sub>O correlation could be caused by nitrous acid (HNO<sub>2</sub>) inhibiting N<sub>2</sub>O reduction as reported by Wicht (1996) (nitrous acid and nitrite equilibrium in solution is pH dependent). Work by Schulthess et al. (1995) has reported that nitric oxide is also an inhibitor of the nitrous oxide reductase enzyme, thereby contributing to N<sub>2</sub>O accumulation.

In contrast to this laboratory research is the findings from several investigations at full-scale facilities. Three studies at wastewater treatment plants (WWTP's) performing biological nitrogen removal (BNR) reported very low N<sub>2</sub>O emissions. Kimochi et al. (1998) reported N<sub>2</sub>O-N conversions between 0.01 and 0.08% of influent nitrogen. This WWTP was modified to attempt to reduce these emissions and they found that by maintaining a dissolved oxygen concentration of over 0.5 mg/L during the aerobic/nitrification stage and allowing 60 min for the anoxic/denitrification stage, that complete nitrification and denitrification with a minimum of nitrous oxide production could be achieved. In fact, their findings state "An optimum combination of aerobic and anoxic conditions and their suitable control are very important for improving nitrogen removal efficiency and controlling N<sub>2</sub>O emissions." At a pilot plant, Thörn and Sörensson (1996), observed an average nitrous oxide production rate of 0.0091 mg N L<sup>-1</sup> h<sup>-1</sup> in the denitrification basin. However, there is insufficient information to convert this value into a percentage of influent nitrogen. Research at a Swiss WWTP with a predenitrification anoxic tank followed by three aerobic tanks also observed a very low nitrous oxide emission (<0.1% of influent nitrogen) (Schulthess and Gujer 1996). However, two studies at secondary treatment facilities (aeration tanks only treating biochemical oxygen demand) have also determined negligible emissions. At the municipal WWTP in Durham, New Hampshire, Czepiel et al. (1995) found N<sub>2</sub>O emissions of only 1.6 × 10<sup>-6</sup> g of N<sub>2</sub>O/L of wastewater or 3.2 g N<sub>2</sub>O/person\*year. Though it is not provided, if one assumes a typical nitrogen content of raw wastewater of 40 mg of N/L (Metcalf and Eddy 1991), this is a N<sub>2</sub>O-N conversion of influent nitrogen of only 0.0025%. Sumer et al. (1995) reported an N<sub>2</sub>O-N conversion of 0.001% of influent N at an activated sludge plant in Germany; however, this plant included a pretrickling filter to increase nitrification in the aeration tank.

If the conversions of reactive nitrogen to N<sub>2</sub>O have been demonstrated by these five scientific studies to be extremely low and therefore of almost no importance, then why discuss it? Unfortunately, the complete picture is much more complicated and uncertain to allow for this simple dismissal. The three BNR plants observed low emissions; it appears that the processes to successfully promote nitrogen removal are consistent with the same processes which ensure complete nitrification and denitrification and thus minimize N<sub>2</sub>O emissions. However, is this representative of all BNR facilities? Kimochi reported that the aerobic/anoxic conditions and their control are very important for controlling emissions. As demonstrated by laboratory research, the possibility exists for poorly operated BNR plants to be important sources of N<sub>2</sub>O emissions. Furthermore, for the two investigations at secondary treatment facilities, there is a distinct possibility that the emissions are not low because the nitrification and denitrification, which occurs in the WWTP, produces little N<sub>2</sub>O. Rather, the emissions are low because it appears that little nitrification or denitrification actually occurred at the secondary treatment plant studied by Czepiel et al. (1995). While nitrification was encouraged at the activated sludge plant studied by Sumer et al. (1995),

there was little or no opportunity for denitrification. Remember, agricultural research has identified denitrification as the more important N<sub>2</sub>O source of the two processes.

An example of past inadequate identification of nitrous oxide emission sources has been demonstrated by a very recent investigation of the South Platte River in Colorado (McMahon and Dennehy 1999). It was found that the N<sub>2</sub>O emissions from this single river, which receives Denver's wastewater effluent, approached the emission estimated by Czepiel et al. (1995) for all the primary plants in the United States. In a much older study, researchers investigating the surface water bodies receiving wastewater effluent from Washington, D.C., found data to imply that "the loss of nitrogen as N<sub>2</sub>O could account for between 2 and 4% of the total nitrogen released by the wastewater treatment plants" (McElroy et al. 1978).

This literature review has demonstrated the following four important points:

- N<sub>2</sub>O emissions can occur from wastewater treatment;
- All or some of these emissions are neglected by not identifying all immediate and future emissions sources;
- Allowing complete nitrification and denitrification will minimize N<sub>2</sub>O formation; and
- Preventing the inhibition of the enzyme nitrous oxide reductase will minimize N<sub>2</sub>O accumulation.

One patent has been filed which reports having developed a "process for reducing nitrous oxide emissions from wastewater treatment" (Spector 1998b). This technology stages both gas and liquid flows (plug flow) so that gaseous or dissolved nitrogen in the form of nitrous oxide, ammonia, and nitrogen oxides are not permitted to exit the system until they have been fully decomposed to N<sub>2</sub> gas. This concept is defined as gas phase staging.

Future research needs to assess the total emissions of nitrous oxide associated with wastewater treatment. If reactive nitrogen remains in the effluent, which is indeed the case with plants performing only primary and secondary treatment, it is important to estimate the future N<sub>2</sub>O emissions from the eventual nitrification and denitrification of this effluent nitrogen in water bodies. Methodology for this has been developed by the IPCC (1997) and emission coefficients were previously presented in the "Background" section. Unfortunately, the variability associated with N<sub>2</sub>O emissions in nature result in a large degree of uncertainty in these emission coefficients. Nevertheless, it is important to begin to estimate the true emissions which will occur and hopefully these coefficients will improve in time. In addition, it is also necessary to quantify gaseous emissions of ammonia and/or nitrogen oxides as potential sources of N<sub>2</sub>O. As previously discussed, these gases are short lived in the atmosphere and are quickly returned to soils or surface water bodies. Once returned to soils or surface water bodies, this reactive nitrogen is again available for nitrification and/or denitrification.

There is also the organic nitrogen contained in the sludge resulting from WWTP which requires assessment as a future N<sub>2</sub>O source. This is typically landfilled, composted (followed by land application), incinerated or directly applied to land. Land application of sewage sludge is directly analogous to applications of chemical fertilizers or animal manures (and well documented by agriculture research). The activities of landfilling, composting and incineration will be discussed further in the following sections.

As a result of the uncertainty around estimating N<sub>2</sub>O emissions from wastewater, the latest guidelines of the IPCC (1997) (and Mosier et al. 1998) advise using an emission coefficient of 0.01 kg N<sub>2</sub>O-N/kg sewage-N produced (it ranges between 0.002 and 0.02 kg N<sub>2</sub>O-N/kg sewage-N produced). This estimate assumes

that 1% of wastewater nitrogen will be emitted as  $N_2O$ -N either immediately or in the future. This estimate will likely be improved upon with new research. The United States has used this emission coefficient as an estimate in the Environmental Protection Agency's latest national greenhouse gas inventory (USEPA 1999). This results in an estimated emission of 27,000 tonnes of  $N_2O$  from 1997 alone and can be converted to 8.4 million tonnes of carbon dioxide equivalent,  $MtCO_2e$  (converted into terms of  $CO_2$  by using the  $N_2O$  global warming potential of 310, as recommended by the IPCC 1995). While it is only a small part of the 6651  $MtCO_2e$  estimated to be emitted by the United States in 1997, it is a more important part of the 931  $MtCO_2e$  nonenergy related greenhouse gas emissions (energy accounts for 86% of total United States greenhouse gas emissions). Furthermore, this may be an underestimate as the estimated nitrous oxide contribution from agriculture is 283  $MtCO_2e$  for the United States in 1997 based on 75% of applied nitrogen. If one was to assume a similar conversion of the 25% carried taken from the farm as food products a far greater result is produced. This waste derived  $N_2O$  could be as high as 94  $MtCO_2e$  (i.e., both liquid and solid forms).

### Landfill Disposal

There were only two research papers found addressing nitrous oxide emissions from landfills. Unfortunately, only one of these studies actually investigated the  $N_2O$  emissions resulting from waste disposed in landfills—the other study examined the emissions from different materials as cover soils. In the Japanese study, two landfills, one active and the other closed, both with over 10 million tonnes of refuse in place, were examined (Tsujiimoto 1994). Average  $N_2O$  emissions of 40.2 and 7.8 g/day for the active and closed landfills, respectively, were observed in the landfill gas vented. This emission, while important [40 g/day of  $N_2O$  will equal the annual greenhouse gas emission of 4.5 tonnes of  $CO_2$  equivalent (using the  $N_2O$  global warming potential of 310) or about the combustion of 1600 L of diesel (using the emission estimate of 2.8 kg of  $CO_2/L$  of diesel from Jaques et al. (1997)), represents only 9.3 kg of  $N_2O$ -N annually or 0.02% of an assumed 50,000 kg of nitrogen in the waste [using a nitrogen content of 0.5% for typical waste (White et al. 1995)]. Either this small emission indicates limited nitrification/denitrification taking place or limited  $N_2O$  leakage of the nitrification/denitrification taking place. Given the anoxic/anaerobic nature of landfills, it is likely the former. The other study, performed in Sweden, determined that about 1.6% of sludge nitrogen used as a landfill cover soil will be emitted as  $N_2O$ -N during the first two years (Börjesson and Svensson 1997). This is similar to the 1.25%  $N_2O$ -N estimate for fertilizer and manure nitrogen found in agriculture research (IPCC 1997) and is not surprising considering that the land application of sludge is analogous to fertilization.

Do appreciable amounts of nitrous oxide eventually become emitted from nitrogenous waste disposed in landfills? The Japanese study has demonstrated the existence of an emission, albeit a small one. Appreciable  $N_2O$  emissions likely occur indirectly via landfill leachate. Reactive nitrogen contained in landfilled organic waste is released when anaerobic decomposition occurs. At this point, nitrogen is free to undergo nitrification and denitrification or to be leached by water percolating through the fill. This nitrogen can dissolve in the percolating water and exit the landfill in the leachate. Landfill leachate is known to be high in nitrogen, with a typical total nitrogen concentration (Organic-N+ $NH_3$ -N+ $NO_3$ -N) between 25 and 1600 mg/L for active landfills and between 105 and 170 mg/L for mature landfills

(Tchobanoglous et al. 1993). In recent years, ammonia concentrations in excess of 2,000 mg/L are even being reported (Henderson and Atwater 1995; Rettenberger 1998; Robinson et al. 1998). Assuming that a minimum of nitrification (and thus little denitrification) occurs inside the landfill because of the anaerobic environment, the likely pathway of  $N_2O$  emissions could be from the treatment of landfill leachate. As a result, the previous section on  $N_2O$  from wastewater is applicable here. Even if leachate did enter groundwater aquifers, there are still IPCC methodology for estimating  $N_2O$  emissions in this manner, as it falls under the methodology for nitrogen lost from farm fields.

There are  $N_2O$  emissions from the landfill disposal of organic waste, such as food scraps, sewage sludge, and yard trimmings (assuming these are probably the only materials with appreciable nitrogen contents which are landfilled), but the nature of a landfill probably causes most of these emissions to be future rather than immediate emissions. As anaerobic decomposition has been known to continue in excess of 50 years, landfilling temporarily delays the nitrification and denitrification which will eventually occur, thereby delaying the  $N_2O$  leakage which will probably occur as well.

The full life-cycle emissions of landfilled organic waste need to be correctly assessed as a start for mitigation to begin.

### Composting

The composting of organic waste will result in  $N_2O$  either during the process or by the future nitrification and denitrification of reactive nitrogen leaving the composting process in the form of ammonia emissions, nitrate or organic matter. The available research has demonstrated the existence of  $N_2O$  emissions during the composting process. This is likely  $N_2O$  leakage during the nitrification and denitrification of reactive N in the organic wastes. Researchers have observed a conversion of N to  $N_2O$  ranging from 0.1 to 2.2% and is summarized in Table 3.

The above research documented immediate  $N_2O$  losses during composting, but this is not the complete picture. Additional  $N_2O$  generation is possible from the reactive nitrogen frequently lost during composting and from the reactive nitrogen contained in finished compost. Nitrogen losses in the form of ammonia occur during composting when C:N ratios are below 20:1 and “the available carbon is fully utilized without stabilizing all of the nitrogen.” (NRAES 1992) In addition, finished compost is typically land applied as a soil conditioner where the reactive nitrogen is free to nitrify and denitrify. As a result, actual  $N_2O$  losses may be greater than demonstrated by these studies.

While research has demonstrated that nitrous oxide results from composting of yard waste, this is not necessarily anthropogenic  $N_2O$ . The global  $N_2O$  cycle which existed in preindustrial times resulted from the  $N_2O$  leakage during the nitrification and denitrification of nitrogen in terrestrial ecosystems. The global climate depended on this greenhouse gas for the radiation balance and the ozone layer was naturally regulated by the production of this ozone depleting substance.

For the  $N_2O$  emissions from yard waste composting to be considered a net greenhouse gas (GHG) emission, it would have to be human-induced emissions. In other words, it would have to be  $N_2O$  emissions which would not have occurred naturally without human interference. There are two possibilities when this could occur (1) yard trimmings which contains anthropogenic N fertilizer (i.e., yard waste resulting from fertilizer applications), and (2) when the human-induced accelerated composting process increases the emission of  $N_2O$  over and above the rotting/

**Table 3.** Review of Nitrous Oxide Emissions from Composting

Compost and method	N <sub>2</sub> O-N loss (% of TKN) <sup>a</sup>	Researcher
Yard waste-turned windrow	0.5	Hellebrand (1998)
Food and yard waste-tunnel and static pile	0.2–0.4	Schenk et al. (1997)
Wastewater sludge and wood ash-aerated static piles	0.7 <sup>b</sup>	Czepiel et al. (1996)
Yard waste-turned windrow	1.2	Ballesterio et al. (1996)
Bedding and horse manure-turned windrow	2.2 (after 60 days)	Ballesterio et al. (1996)
Swine feces and cardboard-aerated and turned in-vessel not given	0.1 0.5–0.8	Kuroda et al. (1996) Hellmann (1995) from Hellebrand (1998)

<sup>a</sup>Total kjeldahl nitrogen (TKN).

<sup>b</sup>Data provided in the research paper was adapted to determine this value.

decomposition of organic waste which would otherwise occur in nature. While it is a simple concept that any N<sub>2</sub>O emission from anthropogenic reactive N should be considered a GHG emission, it may be difficult in reality to differentiate between natural and anthropogenic yard waste nitrogen. It is even more complicated to assess the second possibility.

For the second possibility, the question becomes, “Does composting have any effect on the ratio of N<sub>2</sub>O/NO<sub>3</sub> products of nitrification or any effect on the ratio of N<sub>2</sub>O/N<sub>2</sub> products of denitrification?” If it does, the difference in N<sub>2</sub>O would have to be considered as anthropogenic interference.

As previously discussed, N<sub>2</sub>O leakages during denitrification has been speculated to occur when the availability of nitrate (the electron acceptor) greatly exceeds the available organic carbon (for decomposition) causing the electron acceptor to be incompletely utilized (Firestone and Davidson 1989). This could conceivably occur in the anoxic zones of large composting piles which are high in nitrogen. Due to the anoxic conditions, microorganisms could begin using available nitrate as electron acceptors in order to continue organic matter decomposition. By this method, there is the possibility of human-induced composting increasing N<sub>2</sub>O emissions above what would naturally occur.

Another waste material frequently composted is food scraps, especially in backyard composters, and biosolids (sludge) from wastewater treatment plants. Following from the previous assumption that all of the reactive N in food products is human induced, there needs to be N<sub>2</sub>O accounting from the composting of food-derived wastes. As composting has been demonstrated to

directly result in N<sub>2</sub>O emissions and will conceivably result in future emissions, it follows that the composting of food scraps and wastewater biosolids will cause anthropogenic N<sub>2</sub>O emissions.

The IPCC methodology does not yet consider composting as a N<sub>2</sub>O source; agriculture is the main human-induced source and wastewater treatment is indirectly considered because of the anthropogenic fertilizer component of wastewater. However, it is likely that composting will be included in the future.

### Incineration

Limited research has demonstrated that emissions of nitrous oxide occur during the incineration of municipal solid waste (MSW) and wastewater sludge. This research was compiled by de Soete (1993) for an IPCC Workshop (adapted).

The data presented in Table 4 demonstrates the limited and highly variable research of the N<sub>2</sub>O emissions resulting from waste incineration. Examples of emission estimates for municipal solid waste incineration which are being used are as follows (Jaques et al. 1997; USEPA 1999; USEPA 1998):

- Environment Canada National Inventory 160 g N<sub>2</sub>O/tonne waste incinerated;
- U.S. EPA National Inventory 30 g N<sub>2</sub>O/tonne waste incinerated; and
- U.S. EPA MSW Analysis 130 g N<sub>2</sub>O/tonne waste incinerated.

**Table 4.** Review of Nitrous Oxide Emissions from Incineration

Waste-Facility	Temperature (°C)	N <sub>2</sub> O emission (g N <sub>2</sub> -O/tonne waste)	Researcher <sup>a</sup>
Municipal refuse—stepgrate	780–880	11–43	Yasuda et al. (1992)
Municipal refuse—stepgrate	780–980	40–220	Yasuda et al. (1992)
Municipal refuse—fluidized bed	830–850	14–123	Yasuda et al. (1992)
MSW—5 stokers (20–400 tpd)	not given	26–270	Watanabe et al. (1992)
MSW—3 fluidized bed	not given	97–293	Watanabe et al. (1992)
MSW—rot. kiln (120 tpd)	not given	35–165	Watanabe et al. (1992)
Wastewater sludge—rotary grate	750	227	Yasuda et al. (1992)
Wastewater sludge—fluidized bed	770–812	580–1528	Yasuda et al. (1992)
Wastewater sludge—rotary grate	838–854	684–1508	Yasuda et al. (1992)
Wastewater sludge—rotary grate	834–844	275–886	Yasuda et al. (1992)
Wastewater sludge—rotary grate	853–887	101–307	Yasuda et al. (1992)

<sup>a</sup>As quoted in de Soete (1993)—unable to acquire primary reference

Though there is uncertainty in the estimated  $N_2O$  during waste combustion, the data in Table 4 suggests the following three key points:

1. Both the combustion of solid waste and sludge result in the formation of  $N_2O$ ;
2. Wastewater sludge combustion, which generally has a higher N content than MSW, may result in greater  $N_2O$  emissions; and
3. Increasing the combustion temperature during sludge incineration may decrease  $N_2O$  formation.

Both solid waste and wastewater sludge incineration have been demonstrated to result in the thermal formation of nitrous oxide. While the emissions from MSW incineration range anywhere between 11 and 293 g of  $N_2O$ /tonne of waste, the emissions from wastewater sludge incineration range from 101 to 1528 g of  $N_2O$ /tonne of waste. This may be an indication that sludge, which generally has a higher N content than MSW, may result in greater  $N_2O$  formation and therefore emissions. Is thermal  $N_2O$  formation a function of nitrogen content? Research has also demonstrated  $N_2O$  formation during the fluidized-bed combustion of coal and two excellent review papers on this subject are Johnsson (1994) and Wojtowicz et al. (1994). This is important, because  $N_2O$  formation during coal combustion has been found to “originate mainly from the nitrogen present in the carbonaceous fuel (fuel-N).” (Wojtowicz et al. 1994) Also, recent research on MSW incineration has found a correlation between  $N_2O$  formation and the N content of wastes incinerated (Tanikawa et al. 1995). In addition, experiments with the incineration of sewage sludges and various coals found that the sludges yielded higher  $N_2O$  emissions and it was concluded that this was due to the higher nitrogen contents offering a greater potential for  $N_2O$  formation. Therefore, the formation of  $N_2O$  may likely be the conversion of part of the reactive nitrogen in the combusted material to  $N_2O$  gas.

Does increasing the combustion temperature during sludge incineration cause a reduction in thermal  $N_2O$  formation? While the results of Yasuda (1992) are variable, the  $N_2O$  emission was lowest, 101–307 g/tonne, when the temperature was 853–887°C. An exception to this was the result of 227 g/tonne at a temperature of 750°C. Clearly, the only conclusion that can be derived from these results is that future research is greatly required. However, the decrease in  $N_2O$  formation with an increased combustion temperature has been conclusively demonstrated in coal research (Pels et al. 1994; Wojtowicz et al. 1994). Unfortunately, as  $N_2O$  formation decreases, the formation of NO increases, in fact, the sum of fuel nitrogen conversions “was found to be remarkably constant” over a range of temperatures” (Pels et al. 1994). Nitric oxide is also of environmental concern for its contribution to acid rain and photochemical smog. In contrast, the study by Tanikawa et al. (1995), was unable to find a correlation between  $N_2O$  emission and temperature during MSW incineration, while sludge incineration experiments by Werther et al. (1994), found that increasing the freeboard temperature lead to a decrease in  $N_2O$  without a consequent increasing of  $NO_x$  emissions. It is important to remember that  $NO_x$  is a future source of  $N_2O$  emissions; this reactive nitrogen is short-lived in the lower atmosphere and is returned to terrestrial ecosystems in the form of  $HNO_3$  and  $NO_3$  where it is then available for denitrification. Could solid waste and sludge incineration provide an effective means of performing clean denitrification? If the potential exists, the consequent increases in  $NO_x$  emissions must be contended with.

Current air pollution control technology is sophisticated and is effective at reducing  $NO_x$  emissions to  $N_2$  gas by selective cata-

lytic reduction (SCR) and selective noncatalytic reduction (SNCR)—these technologies are well reviewed by Radojevic (1998). Both these technologies normally operate by injecting ammonia or urea into the flue gases to stimulate  $NO_x$  reduction with SCR also using a catalytic bed to improve performance. Problems with these technologies are the potential for emission of unreacted ammonia, referred to as “ammonia slip,” and for the oxidation of ammonia to  $N_2O$ . Assuming that 1% of  $NO_x$ -N and 1% of  $NH_3$ -N emitted in flue gases to the atmosphere goes on to become  $N_2O$  [current emission factors (IPCC 1997)], there will be a minimum  $N_2O$  emission expressed by the following:

$$\begin{aligned} \text{Min. } N_2O = & \text{Immediate } N_2O_{\text{FORMA}} + 0.01 \times NO_{x\text{UNTREAT}} \\ & + 0.01 \times NH_{3\text{AM. SLIP}} \end{aligned}$$

The questions that need to be answered are the formation rates of  $N_2O$  and  $NO_x$  and the relationship between ammonia slip and  $NO_x$  reduction.

### Relative Importance

While research is now showing that all facets of waste management can contribute nitrous oxide emissions, some activities maybe of greater importance than others. Just by comparing the nitrogen flows between wastewater treatment and solid waste management, this point can be demonstrated. For a fictitious 1,000,000 person residential city and using typical values available from literature (Metcalf and Eddy 1991), the annual mass of nitrogen in wastewater (mass of N = 0.027 lb of N/cap × d) and the annual mass of nitrogen in food waste from kitchen scraps (municipal solid waste generation = 1.6 kg/cap × d; food waste = 14% of MSW by mass; water content = 70% of food waste; nitrogen content = 2.6% of dry food waste) (Tchobanoglous et al. 1993) can be estimated. Albeit these estimates are rough, they indicate that 4,480 tonnes of N annually enters the wastewater treatment side while only 640 tonnes of N (from solid food waste) would enter the solid waste management system. Therefore, wastewater may be a proportionally higher nitrous oxide source. Only future research, including commercial, industrial and other sources, would be able to accurately estimate the true nitrogen flows.

To demonstrate the importance of nitrous oxide emissions as part of the greenhouse gas emissions from various solid waste management activities the results of a recent life-cycle analysis are provided in Table 5 (Barton et al. 1999). This analysis estimated the total greenhouse gas (GHG) emission resulting from the landfilling, incineration, or backyard composting of food waste to determine which operation results in the least GHGs. Five important GHG sources and sinks were identified. These were emissions of  $CO_2$  from the combustion of diesel fuel during collection, transportation and transportation of the wastes (1) Landfilled waste will anaerobically decompose and generate  $CH_4$  gas, (2) GHG emissions result from the portion of  $CH_4$  gas generated but not captured and combusted by landfill gas collection systems. This analysis only estimated  $CH_4$  emissions from 1999 to 2010 and is likely an underestimate as emissions could continue to occur in excess of 50 years. However, food waste disposed in landfills that does not anaerobically decompose serves as a carbon sink, defined as the capture and secure storage of carbon that would otherwise be emitted to the atmosphere, (3) During incineration, the prevention of GHG emissions can occur as the energy from waste combusted replaces the use of fossil fuels, (4) In addition, nitrous oxide emissions result from all of the three

**Table 5.** Greenhouse Gas Analysis of Solid Waste Management Processes

(tCO <sub>2</sub> e/tonne)	Landfill	Incinerator	Backyard composter
Collection and processing	0.018	0.016	0.000
CH <sub>4</sub> emission	0.147	0.005	0.000
N <sub>2</sub> O emission	0.041	0.132	0.049
Energy generation	-0.006	-0.100	0.000
Carbon sequestration	-0.081	0.000	0.000
Total	0.120	0.053	0.049

Note: Adapted from Barton et al. (1999)

operations analyzed, (5) These were estimated from the same review of literature used in this paper. The results in Table 5 are in tonnes of CO<sub>2</sub> equivalent per tonne of food waste (as is [wet] basis).

While methane from a landfill is the single most important emission, nitrous oxide accounts for 20, 86, and 100 % of the GHG emissions from landfilling, incineration, and backyard composting. This demonstrates its importance for future research efforts.

### Call for Research

“In the end, it is only a question of time until all of the N used in agriculture, regardless of its origin (technical, biological, or atmospheric N fixation), eventually is converted by nitrification and denitrification into N<sub>2</sub>, N<sub>2</sub>O, and NO<sub>x</sub> in one or other of the sectors of agriculture/human nutrition/waste and wastewater management, with the exception of the share that is prevented from it by recycling (farmyard manures, sewage sludge)” (Isermann 1994).

There is a tremendous need for research into the 25% or so of agricultural nitrogen which leaves farms in the form of food products. Part of this nitrogen is separated from food products as a result of processing, and therefore is diverted to either wastewater treatment or solid waste management. The nitrogen in consumed food products, after a brief stay in humans, also becomes the responsibility of the waste management system. But again, the interrelationships are important; a large part of sewage sludge is disposed in landfills, but landfill leachate is pumped to wastewater treatment plants.

The potential exists that waste management facilities are significant contributors of greenhouse gases by virtue of N<sub>2</sub>O emissions. This literature review indicates that a great deal of uncertainty exists with respect to emission factors. At the same time, it holds out the possibility of minimization of N<sub>2</sub>O through proper operating conditions.

What is abundantly clear is that a great deal of research is required to accurately define the magnitude of N<sub>2</sub>O emissions. Until there is certainty that a significant problem exists, there is unlikely to be an urgency to refine methods for N<sub>2</sub>O reduction nor an acceptance on the part of waste managers, particularly wastewater, that greenhouse gas emissions are yet another reason to keep them awake at night.

### Conclusions

When the nitrogen in food products leave the farm, it switches responsibility; no longer is it the responsibility of agriculture, the waste management community takes over. Therefore, any imme-

diated or future nitrous oxide emissions during wastewater treatment, composting, incineration, or landfilling must be considered as anthropogenic interference of the global N<sub>2</sub>O cycle, and as such, is a greenhouse gas emission and an emission of an ozone depleting substance. The importance of this fact is only in the early infant stages of development. It is hoped that this paper will hasten and stimulate this development. Specific conclusions of this literature review are the following:

1. Anthropogenic nitrogen from agriculture is contained in wastewater and food waste and this results in anthropogenic nitrous oxide emissions throughout waste management;
2. Research into N<sub>2</sub>O emissions from waste management facilities has identified direct sources but has largely neglected subsequent offsite N<sub>2</sub>O production;
3. While wastewater treatment facilities may receive the highest loads of anthropogenic nitrogen, thus having the greatest potential for N<sub>2</sub>O losses, all processes managing food or food-derived wastes, such as the landfilling, composting, or incineration identified here, are potential sources; and
4. The highly variable and uncertain behavior of N<sub>2</sub>O emissions necessitates a great need for research in this area.

Given the scarcity of the research in this area, the authors would like to bring attention to several studies reviewed for this paper but not mentioned for they were not consistent with the intent of this review. These can be found in Appendix-Additional References.

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