Improvements to the U.S. Wastewater Methane and Nitrous Oxide Emissions Estimates

Elizabeth A. Scheehle
U.S. Environmental Protection Agency, Office of Air and Radiation, Non-CO2 Greenhouse Gases and Sequestration Branch, Washington DC.

Michiel R.J. Doorn
ARCADIS, P.O. Box 13109, Research Triangle Park, NC 27709.

ABSTRACT

Over the past 2 years, EPA's estimates of both nitrous oxide and methane emissions from wastewater treatment in the United States have increased by over 100 percent. This increase has been due to the addition of new industrial categories and several significant methodological improvements, including a factor to account for emissions from septic tanks, and an expanded estimate for nitrous oxide from domestic wastewater treatment plants. These revisions make the U.S. EPA's estimate one of the most comprehensive estimates of greenhouse gas emissions from wastewater. As a result, US wastewater emission estimates for 1990 have increased from 18.3 Tg CO2 Eq. in 1999 to 36.8 Tg CO2 Eq. in 2001. This paper will describe each of the revisions. First, the paper will discuss the methane emission factor for domestic wastewater, including the impact of septic tanks. Secondly, it will report on experience gathered as a result of developing first-time U.S. emission estimates for significant industry categories from annual production data, wastewater composition and treatment data, as well as input from industry experts. Finally, it will detail the effort to develop a comprehensive compounded estimate of nitrous oxide from wastewater, accounting for direct emissions from the plant processes, as well as for previously unaccounted additional nitrogen loadings from garbage disposals, bath and laundry wastewater.

INTRODUCTION

The ability to estimate and track greenhouse gas emissions accurately is an important tool for policy makers. Emission estimates can be used not only for the U.S. national greenhouse gas inventory, but also as input to emissions projections and mitigation cost analysis, and to track progress of voluntary reduction programs. Wastewater handling has previously been considered a minor source of greenhouse gas emissions in the U.S. and many other countries. Now that the U.S. greenhouse gas inventory has become more sophisticated, renewed attention has been given to this category because it represents a remaining uncertainty in the national inventory.

Wastewater from both industrial and domestic sources is often treated to remove soluble organic matter, suspended solids, pathogenic organisms and chemical contaminants. Soluble organic matter is generally removed using biological processes in which microorganisms consume the organic matter for maintenance and growth, resulting in biomass sludge. Microorganisms can perform this biodegradation process under aerobic or anaerobic conditions, the latter condition producing methane (CH_4). During collection and treatment, wastewater may be accidentally or deliberately managed under anaerobic conditions. In addition, the resultant sludge may be further biodegraded under aerobic or anaerobic conditions. Untreated wastewater may also produce methane if contained under anaerobic conditions. The organic content, expressed in terms of either biochemical oxygen demand (BOD) or chemical oxygen demand (COD), determines the methane producing potential of wastewater (IPCC, 1997).

Both domestic and industrial wastewater may also be a source of nitrous oxide emissions. Some industries produce wastewater with significant nitrogen loadings that is discharged to the city sewer, where it mixes with domestic, commercial, and institutional wastewater. Domestic wastewater includes human waste as well as flows from shower drains, sink drains, washing machine effluent, etc. It is transported by a collection system to an on-site or decentralized wastewater treatment (WWT) system, or a centralized WWT system. Decentralized WWT systems are septic systems and package plants. Centralized WWT systems may include a variety of processes, ranging from lagooning to advanced tertiary treatment technology for removing nutrients. After processing, treated effluent may be discharged to a receiving water environment (e.g., river, lake, estuary, etc.), applied to soils, or disposed of below the surface.

Nitrous oxide (N_2O) may be generated during both nitrification and denitrification of the nitrogen present, usually in the form of urea, ammonia, and proteins. These are converted to nitrate via nitrification, an aerobic process converting ammonia-nitrogen into nitrate (NO_3^-). Denitrification occurs under anoxic conditions (without free oxygen), and involves the biological conversion of nitrate into dinitrogen gas (N_2). Nitrous oxide (N_2O) can be an intermediate product of both processes, but is more often associated with denitrification.

EMISSION INVENTORY IMPROVEMENTS

In the previous inventory, methane emissions for 1990 from domestic and industrial wastewater were 11.2 Tg CO_2 Eq. and 0.7 Tg CO_2 Eq. respectively. In this year's inventory, methane emissions for 2001 for domestic and industrial wastewater were 13.9 and 14.5 Tg CO_2 Eq., respectively. This increase is a result of organic growth as well as two significant changes to the estimation methodology. The methane conversion factor (MCF) was increased to account for the methane produced in septic tanks. Secondly, industrial wastewater was included as a source. The text below discusses these changes.

Similarly, the methodology to estimate nitrous oxide emissions was reviewed for completeness and accuracy. The text below, discusses several new sub-sources of domestic wastewater that previously had not been identified. Conforming to IPCC guidelines, nitrous oxide emissions are reported separately as emissions from "Human Sewage." In 1990 and in 2001, nitrous oxide emissions from this source were estimated at 7.1 Tg CO_2 Eq. and 15.3 Tg CO_2 Eq., respectively.

Methane Emission Factor for Domestic Wastewater Accounting for Septic Tanks.

Domestic wastewater methane emissions in the United States are estimated using the default IPCC methodology (IPCC 2000; IPCC 1997). The total population for each year is multiplied by a per capita wastewater BOD production rate to determine total wastewater BOD produced. It is assumed that 0.065 kilograms of wastewater BOD_5 are produced per day per capita and that 16.25 percent of wastewater BOD_5 is anaerobically digested (MCF). This proportion of BOD is then multiplied by an emission factor of 0.6 kg CH_4/kg BOD_5. The MCF was adjusted upward from 15 percent to 16.5 percent for the 2000 estimates. This adjustment represents a significant improvement in the quality of this part of the inventory because it accounts for the BOD that is being treated in septic systems.

Prior to this adjustment, the U.S. Inventory included septic systems by default. The methane conversion factor that was used was 15 percent. However, this earlier 15 percent value was based upon a 1990 estimate that is assumed to account for a fraction of industrial wastewater. With better data on the MCFs of different systems, amount of wastewater treated by each system, and industrial wastewater, EPA revised the MCF value. The new methane conversion factor of 16.5 percent is justified as follows.

Doorn, et al. (1997) assumed that 5 percent of wastewater at centralized wastewater treatment plants degrades anaerobically. This number is based on expert judgment and accounts for potentially anaerobic conditions in sedimentation basins and other sectors of a plant. Doorn and Liles (1999) include a discussion on the operation of septic systems. Essentially, 50 percent of BOD settle out in the tank, which is considered to be anaerobic. The rest is dissolved BOD that passes through to the drainage field, which is considered to be aerobic. Based on this information, it is correct to adopt a MCF of 0.5 for septic systems. In the United States, 25 percent of the population's wastewater is treated in septic systems (NEEDS, 1996). This implies that 75 percent of domestic wastewater degrades under conditions that are 5 percent anaerobic and 25 percent of the population's wastewater degrades under conditions that are 50 percent anaerobic. As equation (1) indicates, this leads to a MCF of 16.5 percent.

Equation (1) Weighted MCF = 0.05×75 percent + 0.50×25 percent = 16.25 percent

Estimating Methane Emissions from Industry

Methane emission estimates from industrial wastewater were included in the Inventory for the first time in 1999, while estimates were refined in 2000 and 2001. A top-down approach was used to identify pertinent industrial sectors (IPCC 2000). The first step consisted of listing the industry sectors that were believed to produce large volumes of organic wastewater. In ensuing steps, this information was combined with organic loadings estimates (in COD), the estimated percentage of wastewater that is treated on-site, and a factor that addresses what fraction of the organic wastewater COD is likely to be susceptible to anaerobic degradation (TA). The TA is a critical factor that often has to be determined by expert judgment. TA varies between 1.0 for anaerobic lagoons and 0.0 for most other (aerobic, chemical, and physical) processes. Some treatment systems may have polishing and/or settling ponds with anaerobic zones or pockets that may produce small amounts of methane. In addition, small amounts of methane gas may leak from anaerobic digesters.

Ten major industry categories that produce wastewater with significant organics loadings were identified and evaluated qualitatively for their potential to emit methane from on-site wastewater treatment operations. In doing so, EPA estimated all factors, not just wastewater output or BOD. From this evaluation it was concluded that in the U.S., the meat & poultry industry is a source for methane emissions due to extensive use of anaerobic lagoons; and that the pulp & paper industry, and the vegetables, fruits & juices industry are likely sources for some wastewater methane because they use either shallow lagoons or settling ponds that may have anaerobic pockets or anaerobic sludge.

<u>Meat and Poultry Processing Industry</u>

The meat and poultry processing industry in the U.S. makes extensive use of anaerobic lagoons in sequence to screening, fat traps and dissolved air flotation. Accordingly, it was estimated that 77 percent of all wastewater organics from this industry degrades anaerobically (EPA, 1997b). Production output for the meat and poultry industry was 38 million metric tons in 2000, obtained from the U.S. Census (2001). EPA (1997b) provides wastewater outflows of 13 (8-18) m^3/ton and an average COD value of 4.1 (2-7) g/liter. These data are based on actual field measurements at several plants and are summarized in Table 1. In 2001, methane emissions from meat and poultry processing were 8.3 Tg CO_2 Eq. Recently, EPA's Office of Water has published new data on meat and poultry processing in a preamble (EPA, 2003) and these data are being reviewed for further refinement of the various factors used to estimate emissions.

Table 1. Methane Emission Estimates and Parameters for Selected Industries in 2001

Parameters	Units	Meat & Poultry	Pulp & Paper	Fruits, Juices, and Vegetables
Annual production	Metric tons x 1000	38,600	134,276	34,344
WW Generation	(m^3/t)	13	85	5.6
BOD	(g/l)	--	0.4	--
COD	(g/l)	4.1	5	5
TA	(percent)	77	10.3	5
CH_4 Emissions	Tg CO_2 Eq./yr	8.3	5.9	0.3

Pulp and Paper Manufacturing

There are approximately 565 pulp and paper manufacturing facilities in the U.S. that produced 144 million metric tons of wood pulp, paper and paperboard (EPA, 1997a; Lockwood-Post, 2001). Wastewater treatment for this category typically includes neutralization, screening, sedimentation, and flotation/hydrocycloning to remove solids (WORLDBANK, 1999; Nemerow, 1991). The most important step is lagooning for storage, settling, and biological treatment (secondary treatment). Approximately 42 percent of soluble organics passes on to secondary treatment (EPA, 1993). Furthermore, it was assumed that 25 percent of the organics in secondary treatment lagoons degrade anaerobically, while 10 percent passes through to be discharged with the effluent.

In developing estimates for this category BOD was used instead of COD, because BOD numbers were deemed more reliable for the U.S. [The emission factor that was used is the default emission factor from IPCC (2000): 0.6 kg CH_4/kg BOD]. The overall wastewater outflow was estimated to be 85 m^3/ton and the average BOD loading entering the secondary treatment lagoons was estimated to be 0.4 gram BOD/liter. (WORLDBANK, 1999; Nemerow, 1991). Table 1 shows that methane emissions from the pulp and paper category were estimated at 5.9 Tg CO_2 Eq. in 2001.

Fruits, Vegetables, and Juices Processing

In the U.S., treatment of wastewater from fruits, vegetables, and juices processing includes screening, coagulation/settling and biological treatment (lagooning), while effluent is typically discharged to the municipal sewer. Nemerow (1991) specifically mentions (partial) anaerobic degradation in the lagoons. It was assumed that this industry is likely to use lagoons that are intended for aerobic operation, but that given the seasonal large loadings the lagoons may be prone to developing limited anaerobic zones and produce CH_4. In addition, some anaerobic lagoons may also be used. Consequently, it was estimated that 5 percent of wastewater organics degrade anaerobically. In 2001, methane emissions from this source category were 0.3 Tg CO_2 Eq. /year, as shown in Table 1.

Developing a Comprehensive Compounded Estimate of Nitrous Oxide from Wastewater

The EPA has been estimating nitrous oxide emissions from human sewage effluent disposal using the IPCC default methodology (IPCC 1997) with one modification to account for nitrogen removed in sewage sludge. There are several issues associated with this methodology, pertaining to completeness. Using a flow chart (Figure 1) additional nitrogen loading sources were identified and reviewed. In addition, direct emissions from wastewater treatment plants are not included in the IPCC methodology but have been included in the revised EPA estimates. The following sections will detail these two issues.

Previously Unaccounted Additional Nitrogen Loadings

The IPCC methodology uses annual, per capita protein consumption (kg/year). This number is likely to underestimate the amount of protein that is being discharged for several reasons. Food

(waste) that is not consumed is often washed down the drain, especially when the home is equipped with a garbage grinder. Also, bath and laundry water can be expected to contribute to nitrogen loadings. Table 2 includes dry weight data of components entering the wastewater stream (unfortunately no detailed Total Kjeldahl Nitrogen (TKN) data were found). As the table indicates, the dry weight of waste that is directly related to protein intake (i.e., feces and urine) only accounts for 0.20 lb/cap/day, whereas the total loading of other organic matter is 0.55 lb/cap/day. Protein "intake" should be increased to account for additional nitrogen entering the wastewater flow from the sources indicated in Table 2 ("Domestic wastes").

Metcalf & Eddy (1991) and Mullick (in Doorn, 1997) provide a range of 40-50 and 20-50 mg TKN/liter for average wastewater from residences, respectively (average 40 mg/l). These estimates include bathwater, laundry, and the use of garbage disposals. According to the NEEDS Survey (1996), the total volume of wastewater generated in the US in 1996 was 32,175 million gallons per day (MGD), serving 189,710,899 people (72 percent of population, not including the septic system users). So, in 1996, the per capita TKN loading was:

$40\ [\text{mg/l}] \times 32{,}175 \times 10^6\ [\text{gal/day}] \times 3.8\ [\text{l/gal}] \times 365 \times 1/(189.7 \times 10^6) \times 10^{-6} = 9.4\ [\text{kg TKN/yr.person}]$

According to the EPA Inventory, the average protein intake in 1996 was 41 kg/year, which can be converted to 6.56 kg N/year using FracNPR). Thus, the total nitrogen discharged from residences per person is a factor of 1.43 (9.4/6.56) higher than the nitrogen from "intake" (feces and urine) only. Consequently, a factor of "1.4" was introduced in the methodology to account for the extra nitrogen discharge from kitchen, bath, and laundry wastes.

Table 2. Estimate of the components of total (dissolved and suspended) solids in wastewater[1]

	Dry weight (lb/capita.day		
Component	**Range**	**Typical**	**Percentage**
Domestic wastes:			
Feces (solids, 23 percent)	0.07–0.15	0.09	9
Urine (solids, 3.7 percent)	0.09–0.15	0.11	11
Total Feces and urine		*0.20*	*20*
Ground food wastes	0.07–0.18	0.10	10
Sinks, baths, laundry, other wash waters	0.13–0.22	0.18	19
Toilet (incl. paper)	0.03–0.06	0.04	4
Total from domestic wastes[2]	0.41–0.80	0.52	54
Industrial wastes:	0.33–0.88	0.44	46

[1] adapted from Metcalf and Eddy (1991), Table 5-3

[2] excluding water softeners

A significant amount of industrial wastewater is discharged into municipal sewers to be treated at the local WWTP. This is not accounted for in the existing IPCC methodology. The type, composition, and quantity of this co-discharged wastewater will vary greatly from municipality to municipality, and few average numbers exist in the literature. Metcalf & Eddy provide an indicative nitrogen loading of 20 – 85 TKN mg/liter for both residential and industrial wastewater. The residential wastewater ranges from Mullick and Metcalf & Eddy (see above) were 40-50 and 20-50 mg TKN/l. Another indication may be obtained from Table 2: industrial wastes contribute 0.44 lb ds/cap/day

compared to 0.52 lb ds/cap/day for domestic wastes. The activity data representing the amount of protein entering the wastewater stream should be increased to account for nitrogen from co-discharged industrial wastewater. Until better data become available, it is recommended to increase the amount of N by the difference between the total nitrogen loading average and the residental nitrogen loading average, which results in 10 mg/l (which is a factor of 1.25) to account for industrial co-discharge.

Consequently, a factor of 1.75 (1.4 x 1.25) was introduced to account for the extra nitrogen discharge from kitchen, bath, and laundry wastes, as well as industrial wastewater that is co-discharged into sewers. (Note that nitrogen from commercial and institutional wastewater is assumed to be covered under the existing methodology, because the nitrogen is expressed on a per-capita basis.)

Direct Emissions from Wastewater Treatment Plant Processes

Emissions from WWTPs are not accounted for in the current IPCC methodology. A new overall emission factor (4 g N_2O/person.year) to estimate N_2O emissions from U.S. municipal WWTPs is introduced. This emission factor is based on a factor of 3.2 g N_2O/person.year (Czepiel, 1995) multiplied by 1.25 to adjust for co-discharged industrial nitrogen. The nitrogen quantity associated with these emissions is calculated by multiplying the N_2O emitted by $^{(2 \times 14)}/_{44}$.

According to the 1996 NEEDS Survey, approximately 43 percent of municipal WWTPs in the U.S. are "secondary treatment" only, which means that they employ primary treatment followed by activated sludge treatment. Activated sludge treatment is an aerobic process that results in the nitrification of ammoniated nitrogen. This does not mean that no denitrification can take place at such plants. For example, some anoxic pockets may exist that are conducive to N_2O generation. Various researches have conducted field tests at WWTPs to develop N_2O emission factors and estimates. For example, N_2O emissions from a conventional secondary (activated sludge) WWTP in Durham, New Hampshire, were measured by Czepiel et al. (1995), who provides a rough emission factor of 3.2 g N_2O per capita per year. This estimate does not include N_2O emissions from sewers and primary treatment. Czepiel describes the wastewater as medium strength, but does not provide information on any industrial, institutional, or commercial wastewater co-discharge. It is likely that the wastewater includes non-excrement components. Czepiel provides a rough national estimate of 1.2 Gg/year. As a comparison, the 1999 N_2O emissions from U.S. human sewage were 26 Gg.

A plant with nitrification/denitrification (N/D) stages typically has a nitrogen removal efficiency in the range of 90 percent. This nitrogen is removed as nitrogen gas (N_2, N_2O and others) and not in the sludge. If, for instance, the plant is 90 percent efficient, 10 percent of N is discharged in the effluent, and 25 percent of nitrogen is removed in the sludge, this would imply that 65 percent of nitrogen is removed as gas. This may constitute significant N_2O emissions. Schön et al. (1993) provides an emission factor of 7 g N_2O per year per person, for plants with N/D, not including industrial wastewater. Schön et al. note that the measured emissions showed very large diurnal and seasonal fluctuations.

It would be helpful to know how much municipal wastewater in the U.S. undergoes N/D, but no statistics seem readily available. Approximately 44 percent of WWTPs in the U.S. employ "advanced" treatment (1996 NEEDS Survey). Unfortunately, the NEEDS Survey does not specify the type of "advanced" treatment, but the Survey (Report 3) does allow for a statewide search to identify individual plants that employ specific treatment technologies, one of which being biological denitrification. To get an indication of the extent of biological denitrification, we used this option to identify plants with denitrification in the four largest U.S. states (California, Texas, New York, and

Florida) and found that only a few plants in these states incorporate denitrification. Assuming that the four aforementioned states are representative of the whole country, it can be concluded that denitrification was not common in the U.S. when the 1996 NEEDS Survey was conducted. However, the Survey does also point out that advanced nutrient reduction (N and phosphorus) was expected to double in the future. When N/D plants become mainstream, they may constitute a significant source of N_2O.

In summary, it is recommended to adopt a new overall emission factor (4 g N_2O/person) to estimate N_2O emissions from municipal WWTPs (activated sludge or secondary). This emission factor is based on the Czepiel emission factor multiplied by 1.25 to allow for co-discharged industrial nitrogen loadings. This emission factor is uncertain. When better estimates of emissions for U.S. N/D plants become available and when N/D is becoming more wide-spread in the country, this emission factor should be adjusted (probably upward).

Improved Methodology

With the modifications described above, N_2O emissions from domestic wastewater were estimated using the IPCC default methodology with modifications (Equation 2).

Equation (2) $$N_2O(s) = (US_{POP} \times 0.75 \times EF_1) + \{[(Protein \times 1.75 \times Frac_{NPR} \times US_{POP}) - N_{WWT} - N_{sludge}] \times EF_2 \times {}^{44}/_{28}\}$$

where,

$N_2O(s)$	= N_2O emissions from domestic wastewater ("human sewage")
US_{POP}	= U.S. population
0.75	= Fraction of population using WWTPs (as opposed to septic systems)
EF_1	= Emission factor (4 g N_2O/person.year)
Protein	= Annual, per capita protein consumption
1.75	= Fraction of non-consumption protein in domestic wastewater
$Frac_{NPR}$	= Fraction of nitrogen in protein
N_{WWT}	= Quantity of wastewater nitrogen removed by WWT processes $[(US_{POP} \times 0.75 \times EF_1) \times {}^{28}/_{44}]$
N_{sludge}	= Quantity of sewage sludge N not entering aquatic environments
EF_2	= Emission factor (kg N_2O-N/kg sewage-N produced)
$({}^{44}/_{28})$	= The molecular weight ratio of N_2O to N_2

CONCLUSIONS

Over the past 2 years, EPA's estimates of both nitrous oxide and methane emissions from wastewater treatment in the United States have increased by over 100 percent. This increase has been due to the addition of several new industrial categories, significant methodological improvements including a factor to account for emissions from septic tanks, as well as an expanded estimate for nitrous oxide from domestic wastewater treatment plants. These revisions make the U.S. EPA's estimate one of the most comprehensive estimates of greenhouse gas emissions from wastewater.

Domestic wastewater methane emissions in the United States are estimated using the default IPCC methodology (IPCC 2000). The MCF was adjusted upward from 15 percent to 16.5 percent for the 2000 estimates. This adjustment represents a significant improvement in the quality of this part of the inventory because it accounts for the BOD that is being treated in septic systems.

Ten major industry categories that produce wastewater with significant organics loadings were identified and evaluated qualitatively for their potential to emit methane from on-site wastewater treatment operations. In doing so, EPA estimated all factors, not just wastewater output or BOD. From this evaluation it was concluded that in the U.S., the meat & poultry industry is a certain source for methane emissions due to extensive use of anaerobic lagoons; and that the pulp & paper industry, and the vegetables, fruits & juices industry are likely sources for some wastewater methane because they use either shallow lagoons or settling ponds that may have anaerobic pockets or anaerobic sludge.

To account for the extra nitrogen discharge from kitchen, bath, and laundry wastes, as well as industrial wastewater that is co-discharged into sewers a factor of "1.75" was introduced in the methodology. Nitrogen from commercial and institutional wastewater is assumed to be covered under the existing methodology, because the nitrogen is expressed on a per-capita basis.)

Also, a new overall emission factor (4 g N_2O/person) was adopted to estimate N_2O emissions from municipal WWTPs (activated sludge or secondary). This emission factor is uncertain. When better estimates of emissions for U.S. N/D plants become available and when N/D is becoming more wide-spread in the country, this emission factor should be adjusted (probably upward).

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Figure 1: Municipal wastewater treatment flows, N pathways and possible N_2O emissions sources