

Emissions Credits: Opportunity To Promote Integrated Nitrogen Management in the Wastewater Sector

James S. Wang,^{*,†,‡} Steven P. Hamburg,[†] Donald E. Pryor,[‡] Kartik Chandran,[§] and Glen T. Daigger^{||}

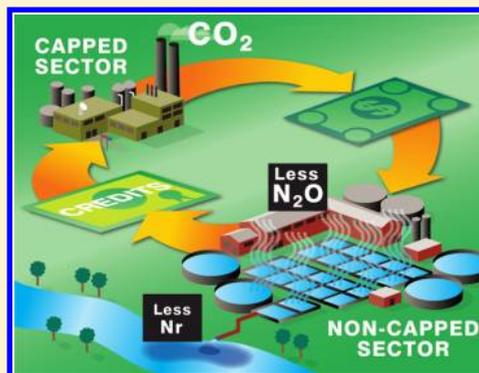
[†]Environmental Defense Fund, 257 Park Avenue South, New York, New York 10010, United States

[‡]Brown University, Providence, Rhode Island 02912, United States

[§]Department of Earth and Environmental Engineering, Columbia University, New York, New York 10027, United States

^{||}CH2M HILL, Englewood, Colorado 80112, United States

ABSTRACT: Relatively little attention has been paid to integrating gaseous N_2O generated by wastewater treatment into overall reactive nitrogen (Nr) pollution reduction. We propose that there is potential for substantial reductions in N_2O emissions through the addition of denitrification processes to existing nitrifying wastewater treatment plants (WWTPs), which are designed to lower ammonia levels but currently do not reduce overall Nr. In addition to providing the benefit of reducing total nitrogen concentrations in the effluent, this kind of WWTP upgrade has been demonstrated to reduce energy consumption and fossil CO_2 emissions. We show that the creation of a greenhouse gas (GHG) crediting system for the wastewater sector could provide a potentially sizable economic incentive on the order of \$10 million to \$600 million per year in the U.S. for upgrading of nitrifying WWTPs that results in N_2O reductions, with an ancillary benefit of another \$30–100 million per year from electricity savings. Even if biological nitrogen removal (BNR) treatment were mandated by existing and future water quality regulations, a GHG crediting system could still be created to promote BNR design and operation that drive N_2O emissions below a baseline to even lower levels. In this case GHG credits could offset around 0.5–70% of the operating and maintenance cost for the BNR.



INTRODUCTION

Human activity has dramatically increased inputs of biologically available nitrogen, or reactive nitrogen (Nr), to the environment, resulting in a host of problems, including worsening air quality and coastal “dead zones”.¹ Increased concern about these negative impacts has led to pollution regulations in many countries, but too often they do little beyond converting Nr from one species to another. In the wastewater sector, one form of “pollution swapping” that occurs at some wastewater treatment plants (WWTPs) is the conversion of ammonia (NH_3) to nitrate (NO_3^-) through the use of microbial nitrification to reduce toxicity. However, conversion of NH_3 to NO_3^- does not by itself constitute biological nitrogen removal (BNR), and WWTPs that include only this process, which we refer to as “advanced secondary treatment” (AST) plants, do not reduce the amount of Nr in effluent entering receiving waters such as rivers and coastal areas.

In 2000, tertiary or BNR treatment designed to remove total Nr from effluent was applied to sewage from only 1% of the 80% of the U.S. population served by centralized WWTPs.² The situation is rapidly changing: Regulators are increasingly requiring BNR treatment to improve local water quality.^{3,4} In brief, BNR employs both nitrification and denitrification processes to convert Nr in the form of NH_3 and organic nitrogen to N_2 gas. However, relatively little attention has been paid thus far to a

byproduct of nitrification and denitrification, N_2O , which is a greenhouse gas (GHG) that is 300 times more potent than CO_2 for a 100-year time horizon.⁵ Also, it has been suggested that N_2O is the most important ozone depleting substance emitted today and possibly for the rest of the century.⁶ Studies have reported a range of N_2O emissions rates from WWTPs, from 0.01%–1.6%⁷ up to as high as 15% of the influent N load.⁸ In addition, some studies have identified certain operating conditions, e.g. nonoptimal aeration and insufficient bioreactor residence times, that may lead to high emissions.^{7,9,10} Thus it is important that the wastewater industry understand, monitor, and minimize N_2O emissions to the extent possible.

In this study, we propose that an upgrade of existing infrastructure with well established technology could go a long way toward reducing WWTP bioreactor N_2O emissions, while simultaneously reducing effluent Nr species, downstream N_2O emissions, and energy consumption. Specifically, by retrofitting existing AST plants to include a denitrification process, N_2O emission rates could potentially be lowered while energy requirements are reduced.^{7,11,12} This assertion arises from the fundamental

Received: February 4, 2011

Accepted: June 15, 2011

Revised: May 26, 2011

Published: July 11, 2011

understanding that denitrifying bacteria can both produce and consume N_2O ,¹³ whereas nitrifying bacteria can produce N_2O but cannot consume N_2O once produced.¹⁴ Field measurements by Ahn et al.⁷ indicate that aerobic zones in WWTPs, where nitrification presumably predominates, are associated with higher N_2O emissions than anoxic zones, where denitrification predominates. In addition, WWTPs that employ concerted action of nitrifying and denitrifying bacteria (for example, simultaneous nitrification-denitrification in oxidation ditches) minimize both liquid-phase N_r and N_2O emissions.⁷ Considering that AST plants treat a sizable proportion of the present-day wastewater flow in the U.S.—around 35% overall according to our best judgment based on industry experience, there is a substantial opportunity to reduce N_2O emissions through retrofits of AST plants at relatively low net cost and with additional environmental benefits.

WWTPs consume chemicals and about 3% of the electricity generated in the U.S.,¹⁵ thus indirectly contributing additional GHG emissions. It is well established that adding denitrification to nitrifying WWTPs can reduce the energy required for a number of reasons. First, the electron accepting capacity of the nitrate generated by nitrifiers can be used by heterotrophic denitrifiers to oxidize organic matter, so that less aeration is needed to oxidize organic matter. Theoretically, around 60% of the energy expended in converting ammonia to nitrate could be recovered.¹¹ Second, denitrification restores about half of the alkalinity that is destroyed in the nitrification process, so that less chemical must be added to provide alkalinity.¹¹ Third, replacing part of the nitrification reactor volume with a denitrification zone reduces the volume that needs to be aerated, although this saves energy only if the nitrification reactor is oversized, which is often the case in our experience. As an example, Rosso and Stenstrom¹² note that WWTPs in Southern California are in fact typically designed with reserve capacity, with the fortuitous consequence that aeration tank volume does not need to be increased to support nitrification-denitrification. A fourth possible reason adding denitrification can save energy is that it removes organic surfactants that would otherwise surround air bubbles during the nitrification stage, thereby helping to improve oxygen transfer efficiency, at least in a study by Rosso and Stenstrom.¹² The resulting decrease in aeration requirement is additional to the lowered oxygen demand due to denitrification. Rosso and Stenstrom reported oxygen transfer efficiency measurements at a number of conventional (secondary), nitrifying-only and nitrification-denitrification WWTPs. They found that nitrification-denitrification systems entail an aeration cost 30% lower than that for nitrifying-only plants.

A promising policy approach to mitigating N_2O emissions from the wastewater sector is linking WWTP design and operation to GHG credits. Under a GHG emissions trading system (cap-and-trade) similar to those that have been considered by the U.S. Congress and the Regional Greenhouse Gas Initiative¹⁶ that is in force for the power sector in the northeast United States, credits or offsets generated by noncapped sectors (e.g., WWTPs) could be purchased by capped sectors (typically industrial CO_2 sources) to comply with emissions limits. We propose that GHG credits could provide an important incentive to upgrade AST plants. Alternatively, when existing and potential future clean water regulations render N removal mandatory, the accompanying N_2O reductions might not be considered “additional,” in the terminology of offsets markets. In other words, standards for offsets generally require that they represent emissions reductions

below what would have occurred in the absence of the emissions reduction incentive or policy. However, even here, the availability of a crediting system could provide an incentive for best practices, if credits are awarded for emissions reductions below a predetermined baseline. The credits could drive N_2O emission factors toward the low end of the observed range and possibly encourage maximum removal of N_r from wastewater at the same time, as discussed below.

The structure of this paper is as follows. After detailing our analysis methods, we present an estimate of the N_2O abatement potential for an upgrade of AST plants across the U.S. and the monetary value of the associated GHG credits (*Part 1: upgrade of AST plants*). We include an estimate of the CO_2 and cost savings associated with the lower energy requirements of BNR WWTPs relative to AST plants. In addition, the sensitivity of the results to values assumed for many of the parameters is discussed. We also describe similar calculations for a scenario in which N removal is mandatory, but GHG credits could provide an incentive to manage N removing plants for maximum environmental benefit (*Part 2: optimizing BNR WWTPs*). We then discuss the evidence for our assertion that reducing N_r in effluent to the lowest levels could simultaneously minimize N_2O emissions. We conclude by discussing additional energy and policy considerations, including how the issue of additionality of emissions reductions from the wastewater sector might play out in the real world.

METHODS

Our study was intended to produce an order-of-magnitude estimate of the value of GHG credits awarded for N_2O emissions reductions achieved at WWTPs to show whether or not a GHG crediting system could provide a sufficient incentive for WWTP managers to adopt efficient N removal processes. In an actual GHG crediting system, the amount of credits awarded would of course depend on the actual performance of individual WWTPs. As such, the study does not employ a detailed emissions model, produce a refined estimate of abatement cost, nor predict industry response to various GHG prices. In addition, we ignore in these calculations methane emissions, an important GHG associated with wastewater treatment, since this paper focuses on management of N_r pollution.

Our methodology for estimating N_2O emissions is based on that of the U.S. Environmental Protection Agency (EPA)² with substantial modifications, which is in turn based on IPCC recommendations.¹⁷ Similar to the EPA methodology, we calculate wastewater emissions as a sum of in-plant and effluent emissions, multiplying together activity data and emission factors (EFs) to estimate emissions for each component. However, rather than considering only two broad categories for WWTPs, non-BNR and BNR (“without nitrification/denitrification” and “with nitrification/denitrification”), we divided WWTPs into four categories with distinct N removal and emissions characteristics: secondary, AST, BNR mid-level, and BNR high-level, the last two representing differing degrees of N removal. (“Mid-level” corresponds to effluent N concentrations of around 10 mg/L, while “high-level” corresponds to around 3–5 mg/L.) This division helps us to assess the effects of upgrading AST plants and of optimizing the performance of BNR plants.

Another major modification of the EPA methodology is that we replaced the person equivalent (PE, 100 gallons/person-day) flow normalized EFs for WWTPs (g N_2O /PE/yr) with EFs derived from the literature expressed as a fraction of either the

Table 1. Model Parameters: General

parameter	symbol	value(s) used	reference
U.S. population (year 2006)		303,000,000	²
fraction of population served by WWTPs		0.79	²
U.S. N in influent--residential, commercial and industrial (kg N/person/yr)	N_{infl}	6.9, 11.7	low value: ref 19 and this study; high value: ref 2
N removal efficiency in sludge (kg N removed/kg N influent)	$F_{\text{rem,slud}}$	0.20	this study
N_2O emission factor in effluent (kg N_2O-N /kg N effluent)	EF_{eff}	0.005, 0.046	low value: ref 17; high value: ref 18
price of GHG credits (USD/ton CO_2 equivalent)	$Price_C$	\$10, \$40	based on projected prices under a cap-and-trade system over the next decade, e.g. ref 20
incremental cost of BNR (USD/kg N removed)	$Cost_{\text{BNR}}$	\$2, \$11	operating and maintenance cost; excludes capital cost low value from ref 21; high value extrapolated from refs 21 and 22 assuming continued future rise in cost.
oxygen equivalent of nitrate denitrified (kg O_2 /kg NO_3^- -N)		2.86	¹¹
efficiency of oxygen transfer (kg O_2 /kWh)		1.2	¹⁹
avoided CO_2 from reduced electricity use (tons CO_2 /kWh)		6.9	²³
price of electricity, U.S. average (USD/kWh)		\$0.06, \$0.10	projected prices under a cap-and-trade system over the next decade; ²⁰ lower end adjusted to reflect lower prices for industrial consumers ²⁴

total nitrogen (TN) in the influent or the TN removed (kg N_2O-N /kg N influent or kg N_2O-N /kg N removed). This allows us to better capture the effects of varying N removal efficiency and varying wastewater flow rates in WWTPs on emissions.

For the effluent EF, the EPA/IPCC default value, 0.005 kg N_2O-N /kg N effluent, is based on an estimate of indirect emissions from agricultural runoff and is possibly an underestimate for more polluted waters.¹⁷ Therefore, we also considered a higher value that we derived from model results (as described below), 0.046 kg N_2O-N /kg N effluent, which still lies within IPCC's large uncertainty range of 0.0005–0.25 kg N_2O-N /kg N effluent.¹⁸ Specifically, we calculated the overall EF for rivers, estuaries, and continental shelves by taking the ratio of the global N_2O emissions in 1990 to the total input of dissolved inorganic N (DIN) to rivers. Similar to the base case in ref 18, we assumed that the input of DIN to rivers is twice the export of DIN by rivers. We also replaced EPA's estimate for the proportion of N removed in sludge, ~9% in 2006, with our own estimate of 20% (Table 1).

To estimate the potential emissions reduction resulting from upgrading AST plants with a retrofit to promote denitrification, we assumed that the entire proportion of U.S. wastewater that is currently treated by AST plants is instead treated by BNR mid-level WWTPs with their lower emission factor (see Table 2). To estimate the potential emissions reduction in a different scenario in which BNR plants are optimized, we compared emissions from "Baseline" and "Optimized" cases, assuming that the "Optimized" case involves a higher N removal efficiency and lower EF than the "Baseline" (Table 2). Our use of the term "baseline" refers to an estimate of minimal compliance if N removal were required and "optimized" refers to what improvements might be possible with incentives to exceed minimal compliance, with GHG credits being awarded for any difference in emissions between the two. We lump together mid- and high-level BNR plants into a single category for the "Baseline" and "Optimized" cases, since we are interested in the effect on emissions and N in effluent when the average EF and N removal efficiency across all BNR plants are improved. Note that we

assume a different partitioning of wastewater flow among the WWTP types under this distinct regulatory regime: lower flows through secondary and AST WWTPs compared to the "Present Day" reflect a mandated switch to BNR treatment. However, we do not expect that regulations would require all AST plants in all watersheds to meet N limits, so we do not assume AST plants are completely eliminated.

We examined the sensitivity of the results by considering plausible ranges for many of the parameters. However, we did not consider all conceivable values for each parameter, since our goal was simply to examine whether N_2O reductions could be financially valuable given improvements in wastewater treatment practices and reasonable parameter assumptions. Low and high estimates are shown in Tables 1 and 2 for the model parameters deemed important for the sensitivity study. One of the assumptions built into these parameter ranges is that the conversion of an AST plant to a BNR plant will lower the in-plant emissions or at worst leave them unchanged; this is supported by the theoretical and experimental studies cited earlier in this paper. We ran the model with every possible combination of low and high values for the different parameters.

RESULTS

Part 1: Upgrade of AST Plants. The N_2O emissions produced by different types of WWTPs and effluent before and after an upgrade to denitrification are depicted in Figure 1. Shown are the two bracketing cases among the numerous model runs associated with the minimum and maximum reductions in total emissions. The emissions reductions for these two cases are also reported in Table 3 as well as the combination of parameter values associated with each. In the minimum case, there is no change in emissions from WWTPs given the equivalence of the EFs for AST and BNR mid-level plants but a small decrease from effluent, as the increase in N removal resulting from the upgrade to BNR reduces the amount of N_r discharged to receiving waters. In the maximum case, there is a sizable decrease in in-plant emissions as a result of $EF_{\text{BNR, mid}}$ being lower than EF_{AST} and a large decrease in effluent

Table 2. Model Parameters: Differences Among WWTP Types^g

parameter	symbol	Upgrade of AST Plants for Denitrification			
		secondary	AST	BNR mid-level	BNR high-level
proportion of U.S. WWTP flow	present day	0.30	0.35	0.30	0.05
	upgrade	0.30	0	0.65	0.05
N removal efficiency to atmosphere (kg N removed/kg N influent)	Frem _{BNRmid}	0	0.10	0.45, 0.60	0.75
	Frem _{BNRhi}				
N ₂ O emission factor in WWTP (kg N ₂ O–N/kg N influent or removed) ^a	EF _{AST}	0.00035 ^b	0.00355 ^c	0.00315,	0.0002 ^f
	EF _{BNRmid}		0.0099 ^d	0.0059 ^e	

parameter	symbol	Optimizing BNR Plants			
		secondary	AST	BNR	
proportion of U.S. WWTP flow		0.20	0.10	0.70	
N removal efficiency to atmosphere (kg N removed/kg N influent)	Frem _{BNRbase}	“Baseline”	0	0.10	0.45, 0.60
	Frem _{BNRopti}	“Optimized”	0	0.10	0.70, 0.80
N ₂ O emission factor in WWTP (kg N ₂ O–N/kg N influent or removed) ^a	EF _{BNRbase}	“Baseline”	0.00035 ^b	0.0099 ^d	0.00315, 0.0059 ^e
		“Optimized”	0.00035 ^b	0.0099 ^d	0.0002 ^f

^a Units are kg N₂O–N/kg N influent for secondary and AST plants, and kg N₂O–N/kg N removed for BNR plants. ^b Reference 25. Equivalent to the value assumed in EPA methodology for WWTPs without nitrification/denitrification, 3.2 g N₂O/PE/yr. ^c Lowest emitting non-BNR WWTP from ref 7, annual average. ^d Highest emitting non-BNR WWTP from ref 7, annual average. ^e Low value: Median of BNR WWTPs from ref 7, annual average. High value: Chosen so that EF for BNR mid-level (expressed relative to influent N) never exceeds that for AST. Note that the EPA EF for WWTPs with nitrification/denitrification, 7.0 g N₂O/PE/yr, lies between those for the lowest- and highest-emitting BNR WWTPs from ref 7, 0.74 and 62.5 g N₂O/PE/yr (annual average). ^f Lowest emitting BNR WWTP from ref 7, annual average. Note that the EPA EF for WWTPs with nitrification/denitrification, 7.0 g N₂O/PE/yr, lies between those for the lowest- and highest-emitting BNR WWTPs from ref 7, 0.74 and 62.5 g N₂O/PE/yr (annual average). ^g Parameter values for which a reference is not indicated are based on the professional judgment of the authors.

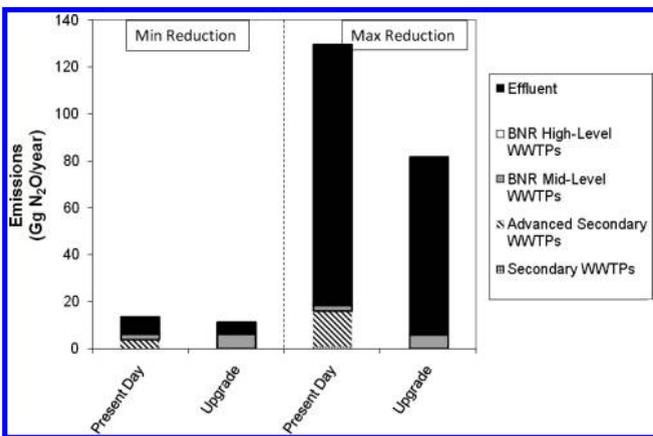


Figure 1. Effect of upgrading AST plants in the U.S. on total wastewater N₂O emissions. Shown are the minimum and maximum emissions reductions out of all of the model parameter combinations tested. (Note that although secondary and BNR high-level WWTPs are included in the legend, their emissions are too low to be visible on this plot scale.)

emissions stemming from the combination of high initial N loading, high N removal in BNR mid-level plants and high EF_{eff}. To put the numbers in perspective, as a result of the upgrades, wastewater N₂O emissions decrease by an amount equivalent to about 0.2% of present-day U.S. total N₂O emissions from all economic sectors for the minimum case and by an equivalent of 4% of U.S. total N₂O emissions for the maximum case.² Note that our estimate for present-day national N₂O emissions from wastewater for the maximum case is much higher than that of EPA, 130 Gg/yr vs 26 Gg/yr for EPA, primarily as a result of our assuming a higher EF for effluent; recalculating EPA’s estimate using the higher EF for effluent

increases their estimate for total wastewater N₂O from 26 Gg to 230 Gg for year 2006.

The reductions in N₂O emissions are potentially worth \$7 million to \$570 million per year, accounting for the range of GHG prices, as shown in Table 3. To put this in perspective, we compared the GHG credit values with the incremental cost of N removal as approximated using the price of N allowances on Connecticut’s Long Island Sound water quality trading market (see Table 1). We find that the GHG credits could offset around 0.2–60% of the cost of the additional N removal (Table 3).

In addition to generating valuable GHG credits, an upgrade of AST plants can reduce energy requirements, as discussed above. Here we provide a simple estimate of the CO₂ and cost savings associated with a reduction in electricity use for aeration. Using the range of N removal amounts from our model runs and values shown in Table 1 for the oxygen equivalent of nitrate denitrified, the efficiency of oxygen transfer driven by electrical blowers, and avoided CO₂ from reduced electricity use, we found that 0.3 to 0.8 million tons of CO₂ could be avoided per year—comparable to the low end of the N₂O reductions in CO₂ equivalents. Assuming a range of projected prices of electricity under a GHG cap (Table 1), the cost savings amount to \$30 million–\$120 million per year, offsetting 1–11% of the N removal cost. Adding this to the revenue from GHG credits results in a total cost offset of around 1–70%. These cost calculations for upgrading AST plants do not account for the additional sources of energy and cost savings discussed above, including reduced alkaline chemical consumption, etc., which would result in even greater cost offsets.

The sensitivity of the results to assumed parameter values are further detailed in Figure 2, which depicts the mean results (N₂O reduction, value of GHG credits, and N removal cost offset) for the low and high values of each parameter. The differences

Table 3. Potential Value of GHG Credits and Energy Savings

	upgrade of AST		"Baseline" to "Optimized"	
	minimum result	maximum result	minimum result	maximum result
net reduction in N ₂ O emissions (Mt CO ₂ Eq./yr)	0.7 ^a	14 ^b	1 ^c	17 ^d
value of GHG reduction (USD/yr)	\$7 million ^e	\$570 million ^f	\$12 million ^g	\$680 million ^h
proportion of O&M cost offset by GHG credits	0.2% ⁱ	60% ^j	0.4% ^k	68% ^l
CO ₂ savings from reduced electricity use for aeration (Mt CO ₂ /yr)	0.3	0.8	--	--
cost savings from reduced electricity use for aeration (USD/yr)	\$30 million	\$120 million	--	--
proportion of O&M cost offset by electricity savings	1%	11%	--	--

^a Combination of parameter values associated with this result: high $Frem_{BNRmid}$, low EF_{AST} , high EF_{BNRmid} , low EF_{eff} , low N_{infl} . ^b Combination of parameter values associated with this result: high $Frem_{BNRmid}$, high EF_{AST} , low EF_{BNRmid} , high EF_{eff} , high N_{infl} . ^c Combination of parameter values associated with this result: high $Frem_{BNRbase}$, low $Frem_{BNRopti}$, low $EF_{BNRbase}$, low EF_{eff} , low N_{infl} . ^d Combination of parameter values associated with this result: The opposite of footnote c, i.e. low $Frem_{BNRbase}$, high $Frem_{BNRopti}$, etc. ^e Same as ^a, plus low $Price_C$. ^f Same as ^b, plus high $Price_C$. ^g Same as ^c, plus low $Price_C$. ^h Same as ^d, plus high $Price_C$. ⁱ Combination of parameter values associated with this result: high $Frem_{BNRmid}$, low EF_{AST} , high EF_{BNRmid} , low EF_{eff} , low $Price_C$, high $Cost_{BNR}$. ^j Combination of parameter values associated with this result: The opposite of footnote i, i.e. low $Frem_{BNRmid}$, high EF_{AST} , etc. ^k Combination of parameter values associated with this result: low $Frem_{BNRbase}$, high $Frem_{BNRopti}$, low $EF_{BNRbase}$, low EF_{eff} , low $Price_C$, high $Cost_{BNR}$. ^l Combination of parameter values associated with this result: The opposite of footnote k, i.e. high $Frem_{BNRbase}$, low $Frem_{BNRopti}$, etc.

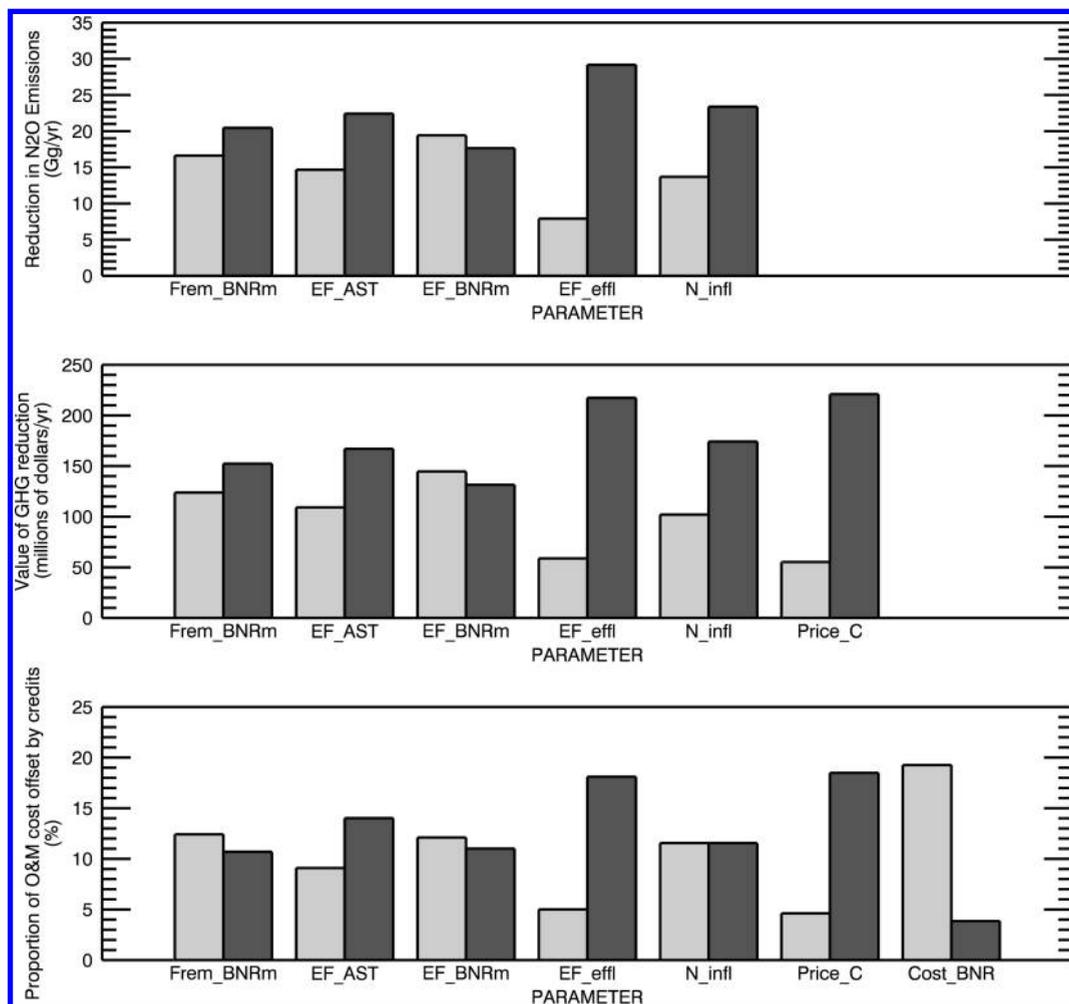


Figure 2. Sensitivity of emissions reduction, value of GHG credits, and BNR cost offset to different parameters, for the upgrade of AST plants scenario. Light gray and dark gray bars represent the mean results for the low and high values of each parameter, respectively. Note that "BNRm" stands for BNR mid-level WWTPs.

between the results for the low and high parameter values indicate the sensitivity, at least for the parameter ranges considered

here. The results are especially dependent on EF_{eff} , $Price_C$, and $Cost_{BNR}$.

The results for N₂O reduction, etc., are independent of the values assumed for $Frem_{slud}$ and $Frem_{BNRhi}$ (not shown).

Part 2: Optimizing BNR WWTPs. The N₂O emissions produced by all WWTPs and effluent for the “Baseline” and “Optimized” cases are depicted in Figure 3. Again, the results associated with the minimum and maximum emissions reductions

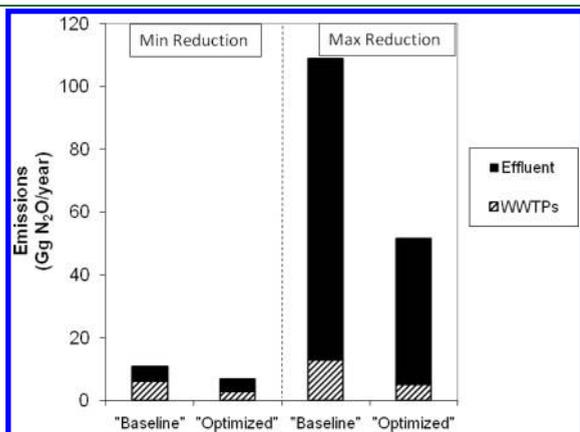


Figure 3. Effect of optimizing BNR WWTPs on N₂O emissions. Shown are the minimum and maximum emissions reductions out of all of the model parameter combinations tested.

are shown. Here emissions from both WWTPs and effluent decrease for both the minimum and maximum results, because of lower in-plant emissions and decreased N_r discharged into receiving waters for the “Optimized” case relative to the “Baseline”.

Note that since the EFs for BNR plants are expressed relative to the amount of N removed, it is the decrease in the product of EF and $Frem$ from the “Baseline” case to the “Optimized” case that results in a decrease in in-plant emissions. We present evidence in the Discussion section that greater N removal could in fact lead to lower total in-plant emissions. Regarding effluent, the decrease in emissions associated with greater N removal is a robust result.

Emissions from wastewater decrease by an equivalent of 0.4–5.0% of present-day U.S. total N₂O emissions. The results in Table 3 indicate that the resulting GHG credits could be worth \$12 million to \$680 million, offsetting on the order of 0.4–70% of operating and maintenance costs of the additional N removal.

Note that in our analysis, we chose a range of values for baseline EF for BNR WWTPs for GHG crediting that happen to be generally lower than the range assumed for EF_{effl} , with a slight overlap, so that the operation of the BNR WWTPs generally reduces total in-plant plus effluent emissions relative to a case in which no BNR is performed and N_r is simply discharged into the environment. However, we recommend that in practice the baseline EF for WWTPs be set significantly lower than the

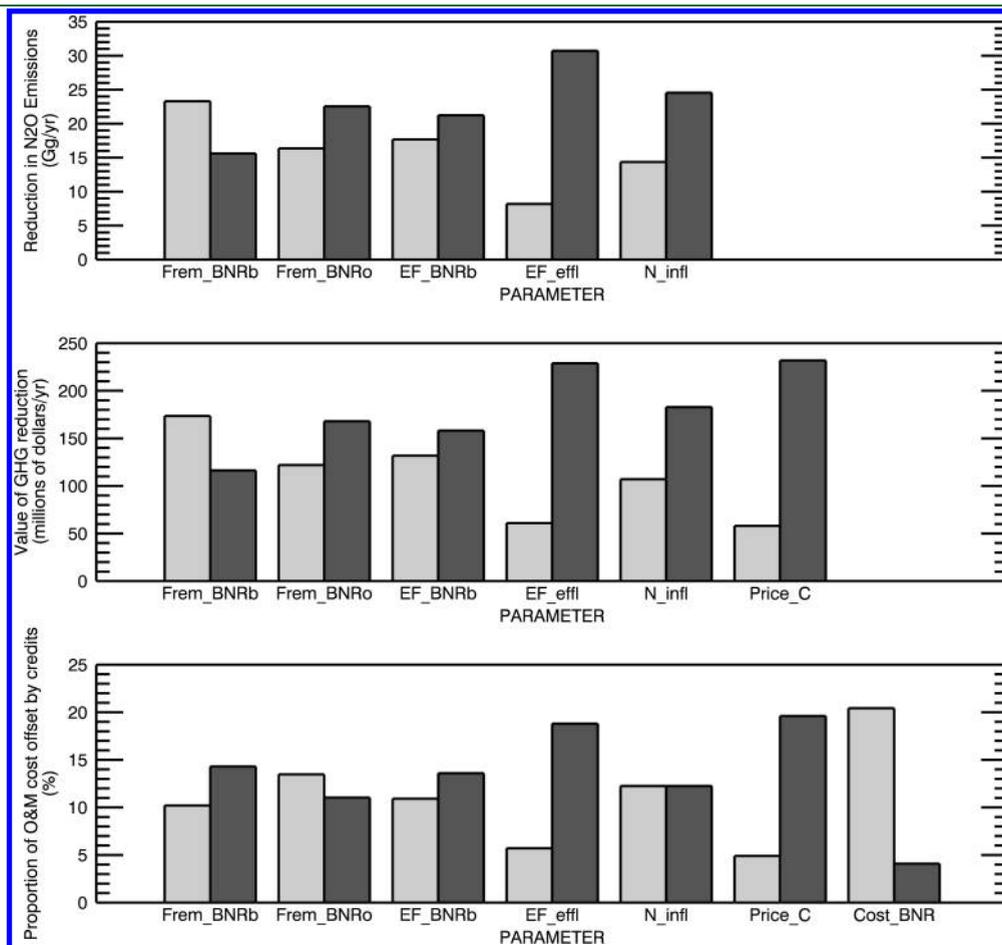


Figure 4. Sensitivity of emissions reduction, value of GHG credits, and BNR cost offset to different parameters, for the optimizing BNR WWTPs scenario. Light gray and dark gray bars represent the mean results for the low and high values of each parameter, respectively. Note that BNRb and BNRo stand for the “Baseline” and “Optimized” cases, respectively.

estimated EF for effluent for a particular location to avoid a perverse situation in which GHG credits are awarded for BNR treatment that actually increases total emissions.

Details of the sensitivities are displayed in Figure 4. The results are especially sensitive to EF_{eff} , $Price_C$, and $Cost_{\text{BNR}}$ for the parameter ranges considered here. The reduction in emissions and their value also depend on the contrast between $Frem_{\text{BNRbase}}$ and $Frem_{\text{BNROpt}}$, i.e. a large improvement in N removal efficiency going from “Baseline” to “Optimized” results in a large reduction in total N_2O emissions. However, since it is more costly to remove more N, the proportion of the N removal cost offset by credits actually decreases the greater the improvement in $Frem$.

DISCUSSION

Relationship between N Removal Efficiency and N_2O Emissions. The outcome of our quantitative example in Part 2, namely, that greater N removal in wastewater treatment processes results in lower N_2O emissions in process reactors (and overall)—a “win-win situation”—is supported by evidence from some recent studies. Foley et al.¹⁰ reported evidence for this in data collected from a number of full-scale BNR WWTPs, suggesting that WWTPs achieving near-complete denitrification emit less N_2O than plants that achieve incomplete N transformation and removal.

The full-scale measurements at a number of WWTPs in the United States by Ahn et al.⁷ provide additional, though indirect, evidence for the relationship between N removal and N_2O . In this study, the authors developed a multivariate regression model to look for significant correlations between N_2O and various operating factors in their extensive data set derived from continuous measurements. The authors also found that N_2O emission fluxes ($g\ N_2O-N/day$) in aerobic zones are positively correlated with concentrations of NH_4^+ , NO_2^- , and dissolved oxygen (DO) and that there is a significant positive interaction between NH_4^+ and NO_2^- . In anoxic zones, there is a significant positive interaction between DO and NO_2^- . These regressions suggest that efficiently running nitrification and denitrification—and thus N removal—with stable DO during the former and low DO during the latter and with little accumulation of NH_4^+ or the inhibitory NO_2^- intermediate results in low emissions (see also ref 26).

In light of the recent findings, it appears that BNR WWTPs that maximize N removal are likely to minimize N_2O emissions at the same time, thereby optimizing the environmental benefit and garnering GHG credits that can offset a portion of the cost of the additional N removal.

Additional Energy Considerations. Adjusting design and operating conditions for nitrification and denitrification to achieve the lowest levels of Nr in effluent and minimize N_2O emissions could potentially consume more energy than moderate levels of N removal, e.g. through mixing an anoxic zone and pumping mixed liquor recirculation, and for some processes the addition of a supplemental organic carbon source such as methanol to promote denitrification, not to mention the possible need for extensive plant retrofits and larger reactor volume.¹¹ In this case, it would be important to encourage the use of renewable energy, including that generated onsite from biosolids (e.g., using an anaerobic digester), to minimize the fossil CO_2 emissions associated with any increase in energy consumption as well as to decrease costs.²⁷ However, some newly developed technologies, such as denitrification and anaerobic ammonia oxidation (anammox), could actually lower the energy consumption

associated with BNR dramatically.²⁸ The right incentives for energy efficiency and N reduction, such as a price on CO_2 and other GHG emissions and Nr discharge and the availability of GHG and N credits, could promote widespread adoption of these improved technologies in the future.

Policy Considerations and Needs. The creation of a GHG crediting system for the wastewater sector could provide an impetus for the addition of denitrification to AST plants, which requires only time-tested and widely used technology and would not only reduce Nr in effluent but also lower N_2O emissions and further reduce GHG emissions through energy savings. The decrease in Nr in effluent thus achieved would be a good first step in alleviating Nr pollution in sensitive watersheds that currently lack N removal requirements for WWTPs, e.g. the Mississippi Basin. Even if BNR treatment were mandated by existing and future water quality regulations, a GHG crediting system could still promote well designed and operated N removal that drives emissions below a baseline to even lower levels.

Whether N_2O emission reductions from the wastewater sector could be considered “additional” might be debated in real-world policy discussions. There are existing policies that provide guidance on this. For example, under the RGGI cap-and-trade agreement among northeastern states, states are advised to employ benchmarks and performance standards to assess the additionality of prospective offset projects.¹⁶ Benchmarks and performance standards are specific approaches to addressing the more general criterion of “financial additionality”. Financial additionality refers to whether or not a project would have been financially attractive in the absence of a crediting system. Performance standards seem particularly relevant in the case of WWTPs. These are quantitative criteria, such as emission rate and market penetration rate, used to determine whether a project surpasses what would occur under standard market practice. Given the low rate of conversion of AST plants to BNR in the U.S. outside of areas covered by N regulations, we believe a strong case could be made for allowing N_2O offsets from unregulated WWTPs into GHG trading markets. Similarly, a case could be made to allow N_2O offsets from regulated WWTPs as long as the emissions are below a certain predetermined level.

“Credit stacking”, in which WWTPs can be awarded credits for simultaneous pollution reductions in multiple environmental trading systems, e.g. water quality and GHG trading systems, are considered controversial by some but could provide additional incentives in dealing with the serious challenge of Nr pollution (e.g., ref 29).

Although the potential N_2O reductions resulting from either AST plant upgrades or BNR plant optimization probably represent <1% of total U.S. GHG emissions,² increased monitoring of N_2O emissions in WWTPs would have the added benefit of providing early warning of nitrification failure.³⁰ We recommend that more WWTPs set up N_2O monitoring to enable the implementation of a GHG crediting system, using a measurement protocol similar to the one reported in ref 7. To date, few studies have measured N_2O emissions from novel N removal processes such as denitrification and anammox at full scale,^{7,31,32} suggesting that further studies are warranted.

AUTHOR INFORMATION

Corresponding Author

*Phone: 301-713-0295. Fax: 301-713-0119. E-mail: James.Wang@noaa.gov.

Present Addresses

[†]NOAA Air Resources Laboratory (R/ARL), 1315 East West Highway, Silver Spring, MD 20910, United States.

ACKNOWLEDGMENT

We thank Michael Oppenheimer, Joseph Rudek, Ramon Alvarez, Gernot Wagner, Arvin Mosier, and anonymous reviewers for manuscript comments. The Table of Contents Art was drawn by Carol Zuber-Mallison. Kartik Chandran's N₂O work was funded by the Water Environment Research Foundation and the National Science Foundation CAREER award.

REFERENCES

- (1) Galloway, J. N.; Aber, J. D.; Erisman, J. W.; Seitzinger, S. P.; Howarth, R. W.; Cowling, E. B.; Cosby, B. J. The nitrogen cascade. *BioScience* **2003**, *53*, 341–356.
- (2) EPA. *Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2008*; 2010.
- (3) EPA. Despite Progress, EPA Needs to Improve Oversight of Wastewater Upgrades in the Chesapeake Bay Watershed; Report No. 08-P-0049; 2008.
- (4) Rhode Island Dept. of Environmental Management. Plan for Managing Nutrient Loadings to Rhode Island Waters, 2005. www.dem.ri.gov/pubs/nutrient.pdf (accessed July 7, 2011).
- (5) Intergovernmental Panel on Climate Change. *Climate Change 2007: The Physical Science Basis—Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*; Solomon, S. et al., Eds.; Cambridge Univ. Press: New York, 2007.
- (6) Ravishankara, A. R.; Daniel, J. S.; Portmann, R. W. Nitrous Oxide (N₂O): The Dominant Ozone-Depleting Substance Emitted in the 21st Century. *Science* **2009**, *326*(5949), 123–125; DOI: 10.1126/science.1176985.
- (7) Ahn, J. H.; Kim, S.; Park, H.; Rahm, B.; Pagilla, K.; Chandran, K. N₂O emissions from activated sludge processes, 2008–2009: Results of a national monitoring survey in the United States. *Environ. Sci. Technol.* **2010**, *44*, 4505–4511.
- (8) Wicht, H.; Beier, M. N₂O emission aus nitrifizierenden und denitrifizierenden Kläranlagen (N₂O emissions from nitrifying and denitrifying sewage treatment plants). *Korresp. Abwasser* **1995**, *42*(3), 404–406, 411–413.
- (9) Kampschreur, M. J.; Temmink, H.; Kleerebezem, R.; Jetten, M. S. M.; van Loosdrecht, M. C. M. Nitrous oxide emission during wastewater treatment. *Water Res.* **2009**, *43*, 4093–4103.
- (10) Foley, J.; de Haas, D.; Yuan, Z.; Lant, P. Nitrous oxide generation in full-scale biological nutrient removal wastewater treatment plants. *Water Res.* **2010**, *44*, 831–844.
- (11) Grady, C. P. L.; Daigger, G. T.; Lim, H. C. *Biological Wastewater Treatment*, 2nd ed.; Marcel Dekker: New York, 1999; 1076 pp.
- (12) Rosso, D.; Stenstrom, M. K. Energy-saving benefits of denitrification. *Environmental Engineer: Applied Research and Practice* **2007**, *3*.
- (13) Zumft, W. G. Cell biology and molecular basis of denitrification. *Microbiol. Mol. Biol. Rev.* **1997**, *61* (4), 533–616.
- (14) Chain, P.; Lamerdin, J.; Larimer, F.; Regala, W.; Lao, V.; Land, M.; Hauser, L.; Hooper, A.; Klotz, M.; Norton, J.; Sayavedra-Soto, L.; Arciero, D.; Hommes, N.; Whittaker, M.; Arp, D. Complete genome sequence of the ammonia-oxidizing bacterium and obligate chemolithoautotroph *Nitrosomonas europaea*. *J. Bacteriol.* **2003**, *185* (9), 2759–2773.
- (15) Kalogo, Y.; Monteith, H. *State of Science Report: Energy and Resource Recovery from Sludge*. Global Water Research Coalition and Water Environment Research Federation. 2008.
- (16) Regional Greenhouse Gas Initiative (RGGI). Overview of RGGI CO₂ Budget Trading Program. 2007. http://rggi.org/docs/program_summary_10_07.pdf (accessed July 7, 2011).
- (17) IPCC. *2006 IPCC Guidelines for National Greenhouse Gas Inventories*, Prepared by the National Greenhouse Gas Inventories Programme, Eggleston, H. S., Buendia, L., Miwa, K., Ngara, T. Tanabe, K. Eds.; Published: IGES, Japan, 2006.
- (18) Seitzinger, S.; Kroeze, C. Global distribution of nitrous oxide production and N inputs in freshwater and coastal marine ecosystems. *Glob. Biogeochem. Cycles* **1998**, *12*, 93–113.
- (19) Tchobanoglous, G.; Burton, F. L.; Stensel, H. D. *Wastewater Engineering: Treatment and Reuse, Metcalf & Eddy, Inc.*, 4th ed.; McGraw-Hill: New York, 2003.
- (20) EPA. *EPA Analysis of the Lieberman-Warner Climate Security Act of 2008*. 2008. http://www.epa.gov/climatechange/downloads/s2191_EPA_Analysis.pdf (accessed July 7, 2011).
- (21) Connecticut Department of Environmental Protection. Report of the Nitrogen Credit Advisory Board To The Joint Standing Environmental Committee Of The General Assembly; 2003. http://www.ct.gov/dep/lib/dep/water/lis_water_quality/nitrogen_control_program/anrpt.pdf (accessed July 7, 2011).
- (22) Connecticut Department of Environmental Protection. Report of the Nitrogen Credit Advisory Board for Calendar Year 2009 To The Joint Standing Environmental Committee Of The General Assembly; 2010. http://www.ct.gov/dep/lib/dep/water/municipal_wastewater/nitrogen_report_2009.pdf (accessed July 7, 2011).
- (23) Green Power Equivalency Calculator Methodologies. <http://www.epa.gov/greenpower/pubs/calcmeth.htm> (accessed July 7, 2011).
- (24) Average Retail Price of Electricity to Ultimate Customers by End-Use Sector, by State. http://www.eia.doe.gov/cneaf/electricity/epm/table5_6_a.html (accessed July 7, 2011).
- (25) Czepiel, P.; Crill, P.; Harriss, R. Nitrous oxide emissions from municipal wastewater treatment. *Environ. Sci. Technol.* **1995**, *29*, 2352–2356.
- (26) Yu, R.; Kampschreur, M.; Van Loosdrecht, M.; Chandran, K. Mechanisms and specific directionality of autotrophic nitrous oxide and nitric oxide generation during transient anoxia. *Environ. Sci. Technol.* **2010**, *44*, 1313–1319.
- (27) Stokes, J. R.; Horvath, A. Supply-chain environmental effects of wastewater utilities. *Environ. Res. Lett.* **2010**, *5*, 014015.
- (28) Kartal, B.; Kuenen, J. G.; van Loosdrecht, M. C. M. Sewage treatment with anammox. *Science* **2010**, *328*, 702–703.
- (29) Credit Stacking. http://my.epri.com/portal/server.pt?open=512&objID=423&&PageID=235467&mode=2&in_hi_userid=230564&cached=true (accessed July 7, 2011).
- (30) Butler, M. D.; Wang, Y. Y.; Cartmell, E.; Stephenson, T. Nitrous oxide emissions for early warning of biological nitrification failure in activated sludge. *Water Res.* **2009**, *43*, 1265–1272.
- (31) Weissenbacher, N.; Takacs, I.; Murthy, S.; Fuerhacker, M.; Wett, B. Gaseous nitrogen and carbon emissions from a full-scale deammonification plant. *Water Environ. Res.* **2010**, *82*, 169.
- (32) Kampschreur, M. J.; van der Star, W. R. L.; Wielders, H. A.; Mulder, J. W.; Jetten, M. S. M.; van Loosdrecht, M. C. M. Dynamics of nitric oxide and nitrous oxide emission during full-scale reject water treatment. *Water Res.* **2008**, *42*, 812–826.