Gaseous Nitrogen Emissions from Anaerobic Swine Lagoons: Ammonia, Nitrous Oxide, and Dinitrogen Gas

Lowry A. Harper,* Ron R. Sharpe, and Tim B. Parkin

ABSTRACT

Seventy-five percent of swine (Sus scrofa) production systems in North America use anaerobic or liquid–slurry systems for waste holding or disposal. Accurate emissions data and emission factors are needed for engineering, planning, and regulatory agencies. These data are used for system design and evaluation of the effect of animal concentrations on the regional soil, surface and ground waters, and atmospheric environments. Noninvasive techniques were used to evaluate trace gases without disturbing the meteorology or lagoon system being measured. Micrometeorological and gas sensors were mounted on a submersible barge in the center of the lagoon for use with flux-gradient methodology to determine trace gas fluxes, without disturbing atmospheric transport processes, over extended periods. Colateral measurements included lagoon nutrient, dissolved gas concentrations, and sludge gas mass flux. Ammonia emissions varied diurnally and seasonally and were highly correlated with wind speed and water temperature. Nutrient loading measurements showed that mobile ions, which were nonvolatile, were constant throughout four successive lagoons. Immobile ions concentrated primarily in the sludge layer of the first lagoon. Measurements of denitrification N losses suggest as much N2-N lost as from NH3-N. Ammonia gas emissions are not as large a percentage of total nitrogen input to the lagoons as previously thought but unaccounted-for nitrogen requires further research.

Ammonia (NH3) is the most abundant alkaline component in the atmosphere that neutralizes sulfur dioxide (SO₂) and N oxides. Consequently, NH₄⁺ is a major component of atmospheric aerosols (particulate matter) and rainfall (Asman, 1994). When NH₄ and NH₃ are deposited onto the soil, nitrification occurs and (in this oxidation process) acid is formed, causing soil acidification (van der Molen et al., 1990) and possible plant nutrient imbalances of Ca, K, and Mg. Ammonia and NH₄⁺ deposition can play an important role in natural ecosystems. Many natural systems such as forest and heath are adapted to low nutrient conditions (Nilsen and Grennfelt, 1988) and when large quantities of N are deposited onto the landscape, nitrophilous species are better competitors. For large parts of northern Europe the N critical loading for many plant species is being exceeded, causing species replacement (Bobbink et al., 1992). In cropping systems, atmospheric NH₃ and NH₄⁺ may be beneficial by adding N during critical times of the day (Harper et al., 1987) and during periods of soil N deficiency (Sharpe et al., 1988; Harper et al., 1996). Crop canopies may also remove significant quantities of NH₃ released to the atmosphere from nearby sources (Harper and Sharpe, 1995; Harper et al., 1996; Bussink et al., 1996).

Increased animal concentrations result in increased efficiency, improved economics, and a better industry support system; however, concentrated livestock production can represent a significant source for NH3 emissions to the atmosphere in a relatively small geographic area. Seventy-five percent of swine production systems in North America use anaerobic or liquid–slurry systems for wastewater, which is a source for NH₃ emissions (Safley et al., 1992b). Ammonia emissions from concentrated animal production sites may influence landscapes and their use (such as forestry, water management areas, fisheries, structural corrosion, aquatic ecosystems) even though the landscape sites may range from a few kilometers to the next state or country from the emission sources. Adverse effects may be due to the direct and indirect effects of NH₃ (which has a shorter residence time of hours) and/or NH₄⁺ aerosols of nitrate (NO₃⁻) and sulfate (SO₄²⁻), which may have a residence time of 5 to 9 d (Crutzen, 1983). Reduction of N losses are also significant economically since the original source was synthetic or biologically fixed N at considerable expense. Providing strong motivation from the animal producer’s viewpoint to promote reduction or prevention of these losses.

In order to evaluate the effect of animal concentration on the local and regional soil, ground water, and atmospheric environments, accurate nutrient dynamics and emissions and emission factors (emissions per animal unit per year) are needed by planning and regulatory agencies. The purpose of this research was to evaluate climate influence on emissions from concentrated swine production on a seasonal and annual basis using noninvasive techniques and to determine the effect of multiple lagoons on nutrient loading and management of the lagoons.

MATERIALS AND METHODS

The swine production unit used in this study is a 12,000 animal facility located in the Coastal Plains of Georgia. The waste disposal system is a series of four lagoons with the animal house waste emptied into the first lagoon. The lagoons are 3.5-, 1.3-, 3.5-, and 1.3-ha for Lagoons 1 through 4, respectively, and the effluent is gravity fed in succession to each of the lagoons. The production facility uses about 300,000 m³ water annually for cleaning and flushing with 82% of the water recycled from Lagoon 4. The balance of the water is from fresh ground water. Effluent from Lagoon 4 (about 29,000 m³

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Abbreviations: TKN, total nitrogen.
is applied to crops and/or pastures through solid-set or center pivot irrigation systems (Sharpe and Harper, 1997). Although trace gases are emitted from all of the lagoons, most of the trace gas measurements for this study were made from the primary lagoon (Lagoon 1).

Micrometeorological instrumentation was located in the center of the lagoon to obtain a minimum fetch of at least 56:1 (upwind lagoon distance to measurement height) in any direction for wind, temperature, and gas concentration profile development. The 50:1 fetch length was marginal in some directions; however, a larger ratio was obtained in the direction of the prevailing winds. Windspeed and NH₃ concentration profile analysis was used to evaluate profile development. Instruments were affixed to a platform “barge” with flotation tanks. The barge with micrometeorological equipment, already mounted and tested, was floated to the center of the lagoon, secured into place with adjustable legs extending to the bottom and guylines attached to the shore. The barge was then sunk to about 50-mm below the water surface to minimize structural interference of windflow patterns by the platform itself. Windspeed (using sensitive cup anemometers, Thornthwaite Associates, Pittsboro, NC), air temperatures (using aspirated thermocouples, Campbell Scientific, Logan UT; Model ASP-TC), and NH₃ concentration profiles were measured above the lagoon at vertical heights of 0.2-, 0.4-, 0.6-, 0.8-, 1.6-, and 2.7-m above the water surface. Ammonia concentrations were obtained by drawing unfiltered air through gas washing bottles containing 80 mL of 0.1 M H₂SO₄, at a known rate (6 L min⁻¹) for 4 h, then transferring it to storage bottles for refrigeration until analysis of NH₃ (by Wards Laboratories, Kearney, NE) concentration by a colorimetric technique similar to that of Weier et al. (1980). Water temperature was measured with thermocouples at three vertical depths, at the surface (about −0.05 m), in the sludge layer (about 3 m), and at a midpoint between the two layers. All electronic data was sampled every minute and averaged over 15 min using a Campbell Scientific Model 21-X data acquisition system.

A schedule of twenty-three, 24- to 48-h data collection periods were made throughout six measurement seasons over the primary lagoon (two each in winter, summer, and spring) to evaluate differences in losses related to micrometeorological and lagoon seasonal variations. Ammonia flux densities were determined during the measurement seasons above the lagoon surface from profile gradients of NH₃ concentrations and the momentum balance transport coefficient.

\[
N = K_{mb} \frac{\Delta n}{\Delta z}
\]

The momentum balance transport coefficient \( K_{mb} \) is determined from the relationship:

\[
K_{mb} = \frac{-k(u_t - u)}{\left( \frac{z_2 - z_1}{z_1} \right)^2 \psi^2}
\]

where \( N = \text{NH}_3 \) flux density (kg ha⁻¹·d⁻¹), \( n = \text{atmospheric NH}_3 \) concentration (μg m⁻³), \( k = \text{von Karman constant}, \mu = \text{windspeed (m s⁻¹)}, z = \text{gradient measurement height (m)}, z_2 = \text{effective vegetation height (m) (note: z_2 is very small over water and was treated as 0),} \) and \( \psi = \text{stability correction factor} \) (no dimensions). This flux-gradient micrometeorological technique was used since it does not interfere with the convective or turbulent transport processes involved in trace gas emissions (Dennemead and Raupach, 1993) and has been used and described previously (Harper et al., 1973; Dennemead et al., 1978). Errors associated with the micrometeorological technique have been discussed by Harper (1988) and Dennemead and Raupach (1992) and error attributed to the technique is ±15%.

Spatial sampling of the lagoon, both horizontally (in three to four random locations) and vertically (at the surface, the vertical midpoint [about 1.5 m], and in the sludge layer), was accomplished using a remotely actuated, closed sampler to obtain samples representative of each of the vertical layers. The sample containers were lowered from a boat to the appropriate depths, opened for sample collection, then closed before bringing them to the surface for sample retrieval and storage. The samples were frozen immediately and shipped to a laboratory (Ward Laboratories, Kearney, NE) for analysis of total nitrogen (TKN), NH₃, NO₃, pH, P, K, S, Ca, Mg, Na, Zn, Fe, Mn, and Cu. For NH₃, a weighed portion of slurry suspension (2 g) was extracted with 2 M KCl. The filtrate was analyzed by flow injection analysis with salicylate-nitroprusside color reagent (Lachat Instruments, Milwaukuee, WI, QuikChem Method 12-107-02-2-A; ammonia [salicylate] in 2 M KCl soil extracts [Anonymous, 1983]). A portion of the slurry suspension was cleared for NO₃-N with calcium oxide (CaO) and analyzed by flow injection analysis by cadmium reduction (Lachat Instruments, Milwaukee, WI, QuikChem Method 10-107-04-1 A, Nitrate/Nitrite, Nitrite in Surface Water, Westwater [Anonymous, 1992a]). For total N, a weighed portion of slurry suspension (about 0.25 g) was placed in a combustion boat with a nickel boat liner and combustion catalyst (tungsten oxide). The combustion boat was then loaded into a Leco (Si. Joseph, MI) FP-2000 N analyzer (Padmore, 1990). For P, K, Ca, Mg, Zn, Fe, Mn, Cu, and Na, a weighed portion of slurry suspension was treated with 10 mL nitric acid. After boiling to reduce the volume to approximately 2 mL, 3 mL of perchloric acid (70%) was added. The solution was heated until it cleared and then was allowed to fume for an additional 20 min. Deionized water was added to the flask while the solution was still hot to bring all minerals into solution. The final volume was brought to 50 mL with deionized water. Phosphorous was determined by developing the metanitrate yellow and reading color intensity with a colorimeter (Anonymous, 1992c). Potassium (Anonymous, 1992b) and sodium (Anonymous, 1992a) were measured by flame emission. Calcium and Mg were determined by atomic absorption after diluting with lanthanum solution (Anonymous, 1992d). Zinc, Fe, Mn, and Cu were measured by atomic absorption (Anonymous, 1992d). The pH of the solution was measured on a 2-g portion of the slurry suspension diluted to 10 mL with deionized water and then read with a glass electrode (Anonymous, 1992d). All lagoons were sampled similarly.

Gas bubbles emitted from each of the lagoons were trapped in three to four collectors in each lagoon below the lagoon surface. The collectors were made of 20-L open-bottom car-
boys (0.275-m diameter) with flotation collars, tethered to the lagoon bottom, for collection of the mass-flow gases (bubbles) before they reached the water–air interface. Gases were collected from the samplers using evacuated sample lines and evacuated SUMA canisters (B.R.C. Rasmussen, Portland, OR). Eight-milliliter gas samples were transferred to evacuated glass autosampler vials capped with butyl rubber stoppers. The samples were analyzed for N\textsubscript{2}, O\textsubscript{2}, and CH\textsubscript{4} using a flame ionization detector (Perkin-Elmer Model 8000, Norwalk, CT) and a molecular sieve column; for CH\textsubscript{4} and N\textsubscript{2} using an electron capture detector (Tracer Model 540, Austin, TX) at 200°C and a molecular sieve column; and for CH\textsubscript{4} and N\textsubscript{2} using a thermal conductivity detector (Tracer Model 540) with the detector at 200°C. Samples were analyzed for NH\textsubscript{3} using an electron capture detector (Shimadzu Mini-2, Columbia, MD) with the detector at 300°C, column temperature of 80°C, a Porapak Q column (Alltech Associates, Deerfield, IL), and N\textsubscript{2} as the carrier gas at 30 mL min\textsuperscript{-1}. Samples were analyzed for CO\textsubscript{2} using an infrared gas analyzer (Model 880A, Rosemount Analytical, La Habra, CA) by passing the effluent from the electron capture detector. Gas fluxes were determined by measuring the amount of gas collected over time and then multiplying the emissions by the measured concentrations.

RESULTS AND DISCUSSION

Ammonia Emissions

Ammonia concentration gradients varied considerably, depending on climatic and lagoon conditions. Figure 1 gives some gradients measured during different seasonal conditions. Windspeeds during the summer period were about 50% higher but there was no difference in Richardson's number (stability). Although a large difference in above-surface concentration is observed between winter and summer, other factors than temperature influence the concentration gradients, including windspeed, water pH, and water nutrient concentration (see below). Normal background NH\textsubscript{3} concentrations varied between 0 and 20 µg m\textsuperscript{-3} (Sharpe and Harper, 1997).

Figure 2 shows the relationship between windspeed and NH\textsubscript{3} flux density during summer 1994 when water temperatures were reasonably constant. Within the summer season, a 40% daily decrease from maximum to minimum windspeeds was followed by a 46% decrease in the NH\textsubscript{3} flux density; however, the small decrease in daily surface water temperature (from 29 to 28°C) could have contributed to a slight decrease in flux density, since colder water results in a reduced gas vapor pressure in the water (about 3%). Figure 3 relates NH\textsubscript{3} emissions to windspeed and water temperature during a period of large weather variation in winter (1996). The general increase in water temperature on day of year (DOY) 45 was due to the passing of a "warm front" moving through the local area. Average daily windspeeds on DOY 43 through 46 were not significantly different, but effluent temperature increased about 10°C during measurement. This increase in temperature resulted in average NH\textsubscript{3} emissions approximately three times larger on DOY 46. During this winter period NH\textsubscript{3} emissions were highly correlated with surface (about 0.05-m deep) water temperature \( r^2 = 0.84 \) but poorly correlated with windspeed \( r^2 = 0.45 \), indicating that the winter season emissions were influenced primarily by water temperature during this time period.

Ammonia emissions were quite variable among seasons (Table 1). The emissions' driving forces (independent effects) were interrelationships of physical and chemical factors. The largest emissions (summer, 1996) occurred during periods of high windspeed and effluent temperature but during relatively low NH\textsubscript{3} effluent concentration. The next highest emissions occurred during periods of moderate windspeeds but higher effluent concentration (spring, 1995). Although winter windspeeds in 1996 were quite high, water temperature was lowest of all measurement periods, resulting in the lowest emissions period. Windspeed and effluent temperature had the highest correlation with NH\textsubscript{3} emissions \( r^2 = 0.86 \) and 0.45, respectively) based on nineteen, 24-h measurements, with minor correlation with effluent concentration and pH \( r^2 = 0.15 \) and 0.13, respectively). The large
Fig. 3. Lagoon ammonia emissions in relation to windspeed (at 1.6-m above the water surface), water temperature (at 0.05-m below the water surface), and to sludge temperature (located within the sludge layer usually around 3.0-m deep, depending on seasonal lagoon storage requirements), Georgia Coastal Plains, winter, 1996.

effect of windspeed on emissions can be illustrated by comparing the partial pressure of NH₃ at the effluent surface during the summers of 1994 and 1996. Although the partial pressure of NH₃ in 1994 was almost twice as high as 1996 (about 0.57 and 0.31 Pa, respectively, and primarily due to a difference in effluent pH), emissions were 12.7 times higher in 1996 with a seasonal average windspeed of 6.7 times higher (Table 1). The low statistical influence of effluent concentration and pH was probably due to small variations in the chemical makeup of the lagoon that resulted from one management system. Combined multiple regression analysis of the six seasons' daily average emissions gave a very good statistical model ($r^2 = 0.94, N = 19$) for emissions on the primary lagoon from this management system, of the form

$$\text{NH}_3 \text{ flux density (kg NH}_3\text{ ha}^{-1}\text{ d}^{-1}) = -275.2817 + \text{windspeed (cm s}^{-1}) \times 0.0510$$

Table 1. Lagoon ammonia emissions and environmental conditions for a swine production system located in the Coastal Plains of Georgia.

<table>
<thead>
<tr>
<th>Season</th>
<th>Year</th>
<th>DOY</th>
<th>Air temperature</th>
<th>Windspeed</th>
<th>Effluent temperature</th>
<th>Effluent NH₃</th>
<th>pH</th>
<th>NH₃ flux</th>
</tr>
</thead>
<tbody>
<tr>
<td>Summer</td>
<td>1994</td>
<td>2420</td>
<td>26.9</td>
<td>168.6</td>
<td>29.5</td>
<td>228.5</td>
<td>8.0</td>
<td>3.88</td>
</tr>
<tr>
<td>Winter</td>
<td>1994</td>
<td>340.5</td>
<td>14.8</td>
<td>159.8</td>
<td>15.4</td>
<td>260.0</td>
<td>7.7</td>
<td>2.35</td>
</tr>
<tr>
<td>Spring</td>
<td>1995</td>
<td>138.3</td>
<td>26.2</td>
<td>286.7</td>
<td>27.8</td>
<td>296.8</td>
<td>7.6</td>
<td>11.93</td>
</tr>
<tr>
<td>Winter</td>
<td>1996</td>
<td>44.5</td>
<td>10.8</td>
<td>470.8</td>
<td>18.3</td>
<td>258.6</td>
<td>7.4</td>
<td>1.56</td>
</tr>
<tr>
<td>Spring</td>
<td>1996</td>
<td>137.7</td>
<td>24.6</td>
<td>191.3</td>
<td>27.1</td>
<td>277.5</td>
<td>7.5</td>
<td>3.79</td>
</tr>
<tr>
<td>Summer</td>
<td>1996</td>
<td>221.5</td>
<td>25.8</td>
<td>1128.5</td>
<td>29.5</td>
<td>237.5</td>
<td>7.7</td>
<td>48.88</td>
</tr>
<tr>
<td>Average</td>
<td></td>
<td></td>
<td>21.4</td>
<td>461.0</td>
<td>23.3</td>
<td>257.2</td>
<td>7.7</td>
<td>12.86</td>
</tr>
<tr>
<td>SD</td>
<td></td>
<td></td>
<td>6.5</td>
<td>342.4</td>
<td>7.6</td>
<td>23.0</td>
<td>0.2</td>
<td>16.81</td>
</tr>
</tbody>
</table>

† At −0.5 m depth.
‡ Day of year.
using 30-min averages gave a poorer estimate ($\rho^2 = 0.61, N = 769$) due to natural diurnal variability of emissions. Although the input variables predict emissions quite well, it should be noted that the model is valid only within the input ranges of Table 1. Further research is needed to expand the predictability of the relationship beyond the narrow range of effluent concentration and pH.

**Mass Flux of Anaerobic Decomposition Gases**

Nitrous oxide ($N\textsubscript{2}O$) emissions, using laser spectrometry and flux-gradient techniques, were measured in the primary lagoon (Lagoon 1) and emissions were not detectable; however, a small, but not significant, absorption of atmospheric nitrous oxide by the lagoon was observed. Further measurements of gases emitted from the sludge layer showed no detectable concentrations of $N\textsubscript{2}O$ (Table 2).

The lack of detectable amounts of $N\textsubscript{2}O$ in solution in the primary lagoon effluent and the low N recovery of input N into the lagoon system suggested N was leaving the production system in other forms. In late spring 1998, gas samples were collected from open-bottom gas collectors and analyzed for $N\textsubscript{2}$, $N\textsubscript{2}O$, $O\textsubscript{2}$, and $CO\textsubscript{2}$. Mass fluxes of the gas bubbles emitted from the sludge were estimated by measuring the collected gas volumes with time through a known cross-sectional area. Table 2 gives measured gas concentrations collected in the four lagoons along with estimated fluxes. Due to the large variability in measurements by the collectors and variability between seasons for both $NH\textsubscript{4}$ and $N\textsubscript{2}$ emissions, there was no significant difference (Tables 1 and 2) between gaseous N emitted as $NH\text sub{4}$ ($12 \pm 17 \text{kg NH}_{4}\text{N ha}^{-1}\text{d}^{-1}$) and as $N\textsubscript{2}$ ($37 \pm 25 \text{kg ha}^{-1}\text{d}^{-1}$) from the lagoon system. (There was large variability in $N\textsubscript{2}$ emissions due to uneven decomposition produced by differential sludge type across the lagoons, from the input to the greatest distance point from the input. The long-term sampling variability in these type studies is about one-third of the mean [Harper and Sharpe, 1998].) Further research is in progress to more closely evaluate mass flux of trace gases during different seasons and management types (L.A. Harper, R.R. Sharpe, F.M. Byers, and W.P. Robarge, unpublished data, 1999).

Results of Table 2 suggest that some form of denitrification is taking place in the lagoons and that different reactions are involved, depending on the N form and concentration. With higher $NH\textsubscript{4}$ concentration and biological activity (i.e., $CH\textsubscript{4}$ production) in the primary lagoon (Lagoon 1) it would appear that denitrification may be chemical denitrification (Van Cleemput, 1997) with minimal $N\textsubscript{2}O$ intermediate loss or biological denitrification by heterotrophs. Thermodynamics and Gibbs free energy for chemical denitrification (Van Cleemput, 1972) suggest that spontaneous conversion of $NH\textsubscript{4}^{+}$ to

Table 2. Mass flux of gases emitted from the sludge layer of a four-cylinder swine waste lagoon system, Carville, LA.

<table>
<thead>
<tr>
<th>Lagoon</th>
<th>$N\textsubscript{2}O$ flux</th>
<th>$N\textsubscript{2}$ flux</th>
<th>$CO\textsubscript{2}$ flux</th>
<th>$CH\textsubscript{4}$ flux</th>
<th>$O\textsubscript{2}$ flux</th>
<th>$NH\textsubscript{4}$ flux</th>
<th>$CO\textsubscript{2}$</th>
<th>$O\textsubscript{2}$</th>
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<tr>
<td>1</td>
<td>15.9</td>
<td>2.1</td>
<td>1.8</td>
<td>0.9</td>
<td>0.4</td>
<td>0.3</td>
<td>15.8</td>
<td>2.0</td>
</tr>
<tr>
<td>2</td>
<td>21.0</td>
<td>3.1</td>
<td>2.2</td>
<td>1.0</td>
<td>0.5</td>
<td>0.3</td>
<td>21.0</td>
<td>3.1</td>
</tr>
<tr>
<td>3</td>
<td>27.0</td>
<td>4.1</td>
<td>3.2</td>
<td>1.1</td>
<td>0.6</td>
<td>0.4</td>
<td>27.0</td>
<td>4.1</td>
</tr>
<tr>
<td>4</td>
<td>33.0</td>
<td>5.1</td>
<td>4.2</td>
<td>1.2</td>
<td>0.7</td>
<td>0.5</td>
<td>33.0</td>
<td>5.1</td>
</tr>
<tr>
<td>Sum</td>
<td>109.0</td>
<td>19.1</td>
<td>16.2</td>
<td>7.3</td>
<td>4.6</td>
<td>3.6</td>
<td>109.0</td>
<td>19.1</td>
</tr>
</tbody>
</table>

* We think the concentrations of $O\textsubscript{2}$ and $CO\textsubscript{2}$ in the mass flux gas analyses are probably influenced by algae growing in the gas collectors.

+ Total nitrogen.
Swine Lagoon, Cordele, GA—Winter and Late Spring, 1996
Effluent Concentrations at Top of Lagoon

Sample locations: 1 = lagoon #1, 2 = lagoon #2, 3 = lagoon #3, 4 = lagoon #4

Fig. 4a. Swine lagoon nutrient concentrations in the effluent (0.05-m below the water surface) and within the sludge layer (approximately 3.0-m deep) in a four-stage lagoon system, Georgia Coastal Plains.

N₂ may occur (O. Van Cleemput and L.A. Harper, unpublished data, 1999). However, it is probable that the process requires an enzyme or catalyst for starting the reaction. Van Cleemput (1998) has shown that in the presence of Fe²⁺ (which is present in the sludge, Fig. 4b [Lb]) and an alkaline pH (Fig. 4a [Ga and Gb]), NO₃⁻ can be chemically reduced in soils. It is possible that there is some biological denitrification in Lagoon 1, but we think it is small since we measure no NO₃⁻ or N₂O, nor do we measure dissolved O₂ necessary for autotrophic nitrification. It is also possible that there are some processes occurring such as nitrification-denitrification by methanotrophs (Verstraete and Philipps, 1998) or by heterotrophs not requiring O₂ but using NO₃⁻ as an electron acceptor (Strous et al., 1997). In lagoons with the highest amounts of methanogenesis occurring, we found the highest quantities of N₂ emitted in relation to other nitorgenous gases (NH₃ and N₂O).
Swine Lagoon, Cordele, GA—Winter and Late Spring, 1996

Effluent Concentrations at Top of Lagoon

<table>
<thead>
<tr>
<th>Conc. (mg L⁻¹)</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
</tr>
</thead>
<tbody>
<tr>
<td>S</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mg</td>
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</tr>
<tr>
<td>Cu</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sample Location</td>
<td>1</td>
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</table>

Sludge Concentrations at Bottom of Lagoon

<table>
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<th>Conc. (mg L⁻¹)</th>
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<th>3</th>
<th>4</th>
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</thead>
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<td>S</td>
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<td></td>
</tr>
<tr>
<td>Mg</td>
<td></td>
<td></td>
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</tr>
<tr>
<td>Na</td>
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<td></td>
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</tr>
<tr>
<td>Zn</td>
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<td>Mn</td>
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<tr>
<td>Cu</td>
<td></td>
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</tbody>
</table>

Sample locations: 1 = lagoon #1, 2 = lagoon #2, 3 = lagoon #3, 4 = lagoon #4

Fig. 4b. Swine lagoon nutrient concentrations in the effluent (0.85-m below the water surface) and within the sludge layer (approximately 3.0-m deep) in a four-stage lagoon system, Georgia Coastal Plains.

(Harper and Sharpe, 1998). Lagoons 3 and 4 had a higher concentration of NO₃⁻ (Fig. 4a [Ca andCb]) and O₂ content in the water surface (stated previously) that may provide conditions more suitable for biological denitrification (Seitzinger, 1990). In these lagoons, a larger percentage of N₂ gas was produced along with significant amounts of N₂O intermediate gas. We think the concentrations of O₂ and CO₂ in the mass flux gas analyses are probably influenced by algae growing in the gas collectors. Further research is needed to evaluate the mechanism of N₂ and N₂O gas production for use in better management possibilities in lagoon waste systems.

Lagoon Nitrogen

Figure 4 shows the comparisons between winter and spring nutrient concentrations in each of the successive lagoons (1 through 4). Average residence times for the effluent were 118 d in Lagoons 1 and 3 and 49 d in 2 and 4. Organic N in the effluent did not change between winter and spring in Lagoon 1 (Fig. 4a [Aa]) but there
was considerable decomposition in the Lagoon 1 sludge layer from winter to spring reflected in the decrease in sludge TKN (Fig. 4a [Ab]) (note: there was little sludge in Lagoons 3 and 4). From winter to spring, the N mass balance in Lagoon 1, including the increase in effluent NH$_4^+$ content from winter to spring, the estimated volatilized NH$_3$ (using the average of winter and spring emissions), the N input from the swine houses, the change in TKN in the sludge, and the N loss to Lagoon 2, indicated about 30% of the total N was not accounted for.

There was no NO$_3^-$ in the input to Lagoon 1 and in the lagoon itself (Fig. 4a [Ca and Cb]). Dissolved oxygen contents further indicated that there was little evidence of nitrification in Lagoon 1 (<0.5 mg L$^{-1}$ O$_2$). Oxygen levels in Lagoons 2 through 4 at the surface indicated nitrification was possible with dissolved O$_2$ contents of 8.7, 18.4, and 20.0 mg O$_2$ L$^{-1}$, respectively, in the top 0.5 m of the lagoons. These concentrations were slightly higher than concentrations found in “clean” water, such as streams, lakes, or ponds, because of O$_2$ produced by algal photosynthesis (Boyd, 1990). Algal growth was observed in Lagoons 2 through 4. Additionally, Table 2 shows increased O$_2$ content as Lagoons 2 through 4 decreased in N content. Likewise, NO$_3^-$ increased with O$_2$ content, probably due to nitrification. Dissolved O$_2$ in the lower layers in Lagoons 2 through 4 was much lower (1 to 2 mg L$^{-1}$ at 1 m) than the surface values in Table 2. It is possible that NO$_3^-$ produced in the surface layers of lagoons was mixed into the anaerobic lower layers (perhaps in the sludge layer) and was denitrified to N$_2$ gas. Our studies of gas bubbles in the water have indicated higher N$_2$ gas concentrations where NO$_3^-$ was present in the lagoon effluent (Table 2). Figure 4a (Ca and Cb) show NO$_3^-$ accumulating during winter in the effluent and sludge in both Lagoons 3 and 4. Figure 4a (Cb) shows a NO$_3^-$ increase in the sludge and lower layers of Lagoons 2 through 4. Nitrification has a lower activity range with respect to temperature than denitrification (Wild et al., 1971; Stanford et al., 1975). Similarly, denitrification activity temperature response is an exponential relationship and nitrification activity follows a Q$^{10}$ factor of approximately 2. Both relative relationships suggest a higher production of denitrification-nitrification in warmer temperatures. We suggest that NO$_3^-$ production in spring, with warmer water temperature, is denitrified to N$_2$ and/or N2O. No N2O was found in the gas collectors in Lagoon 1. Nitrous oxide gradients were measured over Lagoon 1 and were found to be not significantly different from zero. In this lagoon, reduction to N$_2$ was complete without release of a N$_2$O intermediate. Increasing amounts of N$_2$O were measured from Lagoons 2 through 4, along with increasing NO$_3^-$ in the water. We think that increased NO$_3^-$ in solution and N$_2$O produced may be indicative of biological denitrification in these lagoons, particularly Lagoons 3 and 4.

**Lagoon Nutrients**

**Nitrogen**

Ammonium concentrations in the lagoons decreased considerably from Lagoons 1 through 4, decreasing from an annual (winter, spring, and summer) mean in Lagoon 1 of 251 ± 19 mg L$^{-1}$ to 31 ± 19 mg L$^{-1}$ in Lagoon 4.

Irrigated N was determined from pumping records and measured nutrient concentrations and represented a small amount of the total input N. Of the amount applied in irrigation water, Sharpe and Harper (1997) reported 82% of the NH$_4^+$N was lost during application and volatilization after irrigation. They found an additional 13% gaseous loss as N$_2$O within 15 d of application. Similar application losses have been observed by Sallley et al. (1992a). If comparable losses occurred from irrigation during the rest of the year, approximately 0.1% of the total N input into the lagoon system becomes available for crop production in this swine production system.

We suspect part of the unaccounted-for N may have been lost by biological or chemical denitrification in the form of N$_2$ gas from the lagoons. It is possible that NO$_3^-$ and possibly NO$_2^-$ production at the surface (and its diffusion or mass transport to the bottom), along with atmospheric N$_2$O absorption, may provide the electron donor for denitrification (to N$_2$ gas) in this highly reducing condition.

**Other Mobile Nutrients**

Potassium and Na are soluble ions and their concentrations changed little between the lagoons or between effluent and sludge (Fig. 4a [Ea and Eb] and Fig. 4b [Ja and Jb]). Application of irrigation water to the crops represents 912 and 182 kg ha$^{-1}$yr$^{-1}$ of K and Na, respectively. After 5 yr of irrigation, concentrations in the soil profile for K and Na were significantly larger than surrounding nonirrigated pasture (Fig. 5).

**Immobile Nutrients**

Immobile nutrients P, Ca, S, Mg, Zn, Fe, Mn, and Cu show little change in concentration in the effluent from Lagoons 1 through 4 (Fig. 4). However, during winter there is a considerable concentration increase in the sludge layer of the primary lagoon (Lagoon 1). We measured increases in concentration in the sludge ranging from seven times higher than effluent concentrations for S (Fig. 4b [Hb]) to 278 times higher for Zn (Fig. 4b [Mn]).
Further evidence of chemical precipitation was observed in the form of crystals attached to the instrument barge and ropes. Analysis of the crystals gave concentrations of P (11.7%), Mg (8.9%), and Ca (1.5%), suggesting crystallization of struvite (MgNH₄PO₄·6H₂O) and calcite (CaCO₃), as found in other studies by Sommer and Hustvedt (1995). Calcite and struvite will be present as stable solid phases at pH > 7 and 6, respectively, and our effluent and sludge pH did not fall below 7 in these studies. A decrease in sludge concentration occurred during spring with increase in water temperature from the resulting temperature effect on solution chemistry (Fig. 4). Also, increased temperature promotes an increased decomposition rate of organic matter which was deposited during the winter.

Application of irrigation water to the crops represented application of 110 kg P ha⁻¹ yr⁻¹, 117 kg Ca ha⁻¹ yr⁻¹, 76 kg S ha⁻¹ yr⁻¹, 73 kg Mg ha⁻¹ yr⁻¹, 0.1 kg Zn ha⁻¹ yr⁻¹, 2.3 kg Fe ha⁻¹ yr⁻¹, 0.02 kg Mn ha⁻¹ yr⁻¹, and 0.2 kg Cu ha⁻¹ yr⁻¹. Following 5 yr of irrigation, Mg increased significantly (α = 0.95) in the soil horizon from 0 down to 0.6 m but no other nutrients were measured to be significantly higher. Interestingly, Mn significantly decreased in the 0- to 0.3-m soil layer and appeared to decrease in the 0.3- to 0.6-m layer, although due to high variability it was not significant. The soil pH increased significantly on the application sites by one pH unit down to 0.6 m in the soil.

Emission Factors

Emission factors, generally defined in units of kg NH₃ animal⁻¹ yr⁻¹, are used by planners and others to evaluate potential area sources due to concentrated animal production. Emission factors must be used with caution because of variability induced by geography and meteorology, methodology for measurement, type and weight of animals, N content of feedstuffs, housing and management, and, indeed, the lack of use of a standard weight or animal type comparison. Emission factors are generally based on the emissions of a category of swine divided by the number of animals in that category. Asman (1992) developed a composite factor for Europe of 8.5 kg NH₃ animal⁻¹ yr⁻¹ and Battye et al. (1994), using emissions from Europe and USDA Agricultural Statistics Service animal classifications, developed a similar composite factor for the USA of 9.2 kg NH₃ animal⁻¹ yr⁻¹. The lagoon emission factor for the lagoon-to-finish (FF) unit in this study is considerably smaller (2.3 kg NH₃ animal⁻¹ yr⁻¹) than the above factors, but it does not include housing emissions that would increase our factor. However, even adding housing losses, preliminary studies suggest that our emission factors will be less than the European and estimated U.S. factors. Further studies are in progress to evaluate NH₃ (L.A. Harper and R.R. Sharpe, unpublished data, 1999) and CH₄ (R.R. Sharpe and L.A. Harper, unpublished data, 1999) emissions from swine production houses.

In conclusion, this study shows that the use of lagoons for waste management may have less environmental impact due to smaller atmospheric emissions of NH₃ than originally thought, with much of the previously estimated N loss from lagoons released to the atmosphere as N₂ gas. A more critical analysis is needed to better understand the N transformations that occur in such complex biological and chemical systems.

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