Reuse of Concentrated Animal Feeding Operation Wastewater on Agricultural Lands

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Concentrated animal feeding operations (CAFOs) generate large volumes of manure and manure-contaminated wash and runoff water. When applied to land at agronomic rates, CAFO wastewater has the potential to be a valuable fertilizer and soil amendment that can improve the physical condition of the soil for plant growth and reduce the demand for high quality water resources. However, excess amounts of nutrients, heavy metals, salts, pathogenic microorganisms, and pharmaceutically active compounds (antibiotics and hormones) in CAFO wastewater can adversely impact soil and water quality. The USEPA currently requires that application of CAFO wastes to agricultural lands follow an approved nutrient management plan (NMP). A NMP is a design document that sets rates for waste application to meet the water and nutrient requirements of the selected crops and soil types, and is typically written so as to be protective of surface water resources. The tacit assumption is that a well-designed and executed NMP ensures that all lagoon water contaminants are taken up or degraded in the root zone, so that ground water is inherently protected. The validity of this assumption for all lagoon water contaminants has not yet been thoroughly studied. This review paper discusses our current level of understanding on the environmental impact and sustainability of CAFO wastewater reuse. Specifically, we address the source, composition, application practices, environmental issues, transport pathways, and potential treatments that are associated with the reuse of CAFO wastewater on agricultural lands.

New technological innovations and the economic advantage of size have driven a structural shift from small to large concentrated animal feeding operations (CAFOs) in the United States. It has been estimated that confined livestock and poultry animals in the United States generate about 453 million Mg of manure annually (Kellogg et al., 2000; USEPA, 2003). Water use at CAFOs includes drinking water for animals and water used in cooling facilities, sanitation and wash down of facilities, and animal waste-disposal systems. The United States Geological Survey estimated that livestock water use accounted for nearly 1% of the total freshwater withdrawals (excluding thermoelectric power) in the United States (Hutson et al., 2004). Daily use of water for farm animals was reported to be 159 L d^{-1} for a milking cow, 18 L d^{-1} for a hog, and 18 L d^{-1} for 100 chickens. Large volumes of animal manure-containing wastewater, wash water, and storm water runoff can be generated at CAFOs (USEPA, 2001). This manure-contaminated water is typically collected and stored in wastewater lagoons on farms (USEPA, 2001).

Transportation, storage, and treatment of manure and manure-contaminated water are costly (Gleick, 2000). The large volume of waste generated, and the lack of disposal area at CAFOs, further limits the ability for effective manure management. Manure and wastewater are, therefore, usually land-applied within about 16 km of CAFO facilities. When applied to agricultural lands at agronomic rates, manure and wastewater can be a valuable fertilizer and soil amendment that can improve the physical condition of the soil for plant growth (Jokela, 1992; Kapkiyai et al., 1999), reduce power required for tillage (Sommerfeldt and Chang, 1985, 1987), and increase the organic content of soil (Sommerfeldt et al., 1988). The reuse of CAFO wastewater in irrigated agriculture also provides a potential means to reduce the demand for high quality water that is a scarce natural resource in many arid and semiarid regions (Pimentel et al., 2004). Conversely, CAFO manure and wastewater have also been reported to pose a potential risk to environmental resources and to human health (Thorne, 2007; Burkholder et al., 2007). Contaminants of potential concern in

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Abbreviations: CAFO, concentrated animal feeding operation; EC, electrical conductivity; EDC, endocrine-disrupting chemical; ET, evapotranspiration; FC, fecal coliform; NMP, nutrient management plan; ORP, oxidation–reduction potentials; PET, potential evapotranspiration; TKN, total Kjeldahl nitrogen; TOC, total organic carbon; TP, total phosphorus; and TSS, total suspended solids.

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Table 1. A summary of measured total suspended solids (TSS), electrical conductivity (EC), oxidation–reduction potentials (ORP), ammonium (NH_4-N), total Kjeldahl nitrogen (TKN), total phosphorus (TP), total organic carbon (TOC), potassium (K), copper (Cu), and zinc (Zn) concentrations from several swine, poultry, dairy, and beef lagoon water samples (mean with standard deviation, three locations for each lagoon).

CAFO† type	TSS	EC	ORP	NH_4–N	TKN	TP	TOC	K	Cu	Zn
	mg L^{-1}	dS m^{-1}	mV	mg L^{-1}						
Beef feedlot‡	212 ± 28	2.6 ± 0.0	73 ± 26	33 ± 3	63 ± 1	14 ± 0	155 ± 6	277 ± 2	0.02 ± 0.00	0.41 ± 0.45
Dairy‡	718 ± 150	3.1 ± 0.0	−277 ± 24	84 ± 12	185 ± 11	30 ± 1	576 ± 94	178 ± 29	0.66 ± 0.06	0.42 ± 0.05
Poultry§	847 ± 15	11.3 ± 0.2	−331 ± 21	656 ± 34	802 ± 7	50 ± 1	1050 ± 40	1430 ± 200	1.65 ± 0.02	0.75 ± 0.08
Poultry‡	865 ± 79	8.0 ± 0.0	−337 ± 2	289 ± 11	407 ± 2	23 ± 0	374 ± 15	1490 ± 30	0.05 ± 0.00	0.27 ± 0.02
Poultry¶	253 ± 75	4.4 ± 0.0	207 ± 28	58 ± 5	96 ± 0	30 ± 0	114 ± 15	811 ± 14	0.01 ± 0.00	0.08 ± 0.00
Swine sow§	1230 ± 30	12.6 ± 0.0	−348 ± 16	944 ± 23	1290 ± 15	264 ± 5	944 ± 304	137 ± 2	0.38 ± 0.01	15.2 ± 0.14
Swine finisher§	5310 ± 2430	19.4 ± 0.0	−368 ± 2	1630 ± 20	2430 ± 40	324 ± 14	4780 ± 280	242 ± 3	6.87 ± 4.60	10.5 ± 0.58
Swine nursery§	4220 ± 1940	21.5 ± 0.1	−368 ± 5	1370 ± 90	2040 ± 60	368 ± 35	1440 ± 130	4150 ± 210	13.3 ± 2.1	109 ± 21

† CAFO, concentrated animal feeding operation.

‡ Secondary lagoon.

§ Primary lagoon.

¶ Tertiary lagoon.

animal wastes include excess amounts of nutrients (Jongbloed and Lenis, 1998; Mallin, 2000), salts (Chang and Entz, 1996; Hao and Chang, 2003), organics rich in biochemical oxygen-demanding material (Webb and Archer, 1994), heavy metals (Barker and Zublena, 1995), microbial pathogens (Gerba and Smith, 2005; Schets et al., 2005), antibiotics (Shore et al., 1988, 1995; Nichols et al., 1997; Peterson et al., 2000), and natural and synthetic hormones (Hanselman et al., 2003; Raman et al., 2004).

This review contains three sections, namely: (i) Environmental Contaminants; (ii) Land Application; and (iii) Treatments. The Environmental Contaminants section reviews the various contaminants that are found in CAFO lagoon water and why they may pose a risk to the environment and/or human health. The Land Application section reviews the current regulatory framework for land application of CAFO wastewater that is based on Nutrient Management Plans (NMPs), the implicit assumptions and possible weaknesses in NMP design and application, illustrative contaminant loading rates at NMP sites, and potential transport pathways for various lagoon water contaminants. Finally, the section on Treatments discusses potential best management practices and lagoon water treatments that may be needed before land application of CAFO wastewater to minimize risks and dissemination of contaminants in the environment.

Table 2. Indicator microbial populations in whole lagoon samples from different CAFOs (mean with standard deviation, three locations for each lagoon).

CAFO† type	Total coliforms	Fecal coliforms	Fecal enterococci
	cfu per 100 mL		
Beef feedlot‡	3.89E03 ± 0.99E03	1.57E03 ± 0.28E03	2.94E03 ± 1.07E03
Dairy‡	1.91E07 ± 0.16E07	1.08E06 ± 0.11E06	1.53E05 ± 0.23E05
Poultry§	4.73E04 ± 2.09E04	2.91E04 ± 1.07E04	1.41E05 ± 0.67E05
Poultry‡	2.42E03 ± 0.00E03	2.42E03 ± 0.00E03	8.51E04 ± 2.75E04
Poultry¶	5.83E04 ± 0.28E04	2.03E01 ± 0.06E01	2.68E03 ± 1.24E03
Swine sow§	4.89E05 ± 0.70E05	3.34E05 ± 0.43E05	3.69E05 ± 0.39E05
Swine finisher§	1.02E06 ± 0.12E06	9.52E05 ± 3.44E05	8.50E05 ± 4.63E05
Swine nursery§	1.87E05 ± 0.95E05	7.93E04 ± 8.30E04	2.43E05 ± 0.00E05

† CAFO, concentrated animal feeding operation.

‡ Secondary lagoon.

§ Primary lagoon.

¶ Tertiary lagoon.

Review

Environmental Contaminants

Six classes of potential lagoon water contaminants are considered in this section, namely: (i) nutrients and organics, (ii) heavy metals, (iii) salts, (iv) pathogens, (v) antibiotics, and (vi) hormones. Tables 1 to 4 provide illustrative examples of the various contaminant concentrations that were measured in CAFO lagoon water from several farms. Considerable variability is expected in the concentration of these contaminants at different farms due to differences in animal and waste management practices. Hence, the concentration values provided in these tables should be viewed as only reflecting site specific conditions at the indicated farm, and general trends should not be ascribed to these data. Wastewater from these lagoons, however, is actually used for land application at these farms, and therefore these data can be used to provide an estimate of potential environmental concentrations for the various contaminants.

Table 1 provides a summary of measured total suspended solids (TSS), electrical conductivity (EC), oxidation–reduction potentials (ORP), ammonium (NH_4–N), total Kjeldahl nitrogen (TKN), total phosphorus (TP), total organic carbon (TOC), and potassium (K) concentrations from several swine, poultry, dairy, and beef lagoon water samples (Hutchins et al., 2007). Concentrations of heavy metals (zinc and copper), indicator microorganisms, antibiotics, and estrogen hormones in these lagoon water samples are also provided in Tables 1, 2, 3, and 4, respectively. For these data, separate analyses were conducted on the samples collected as described previously (Hutchins et al., 2007). Heavy metals were analyzed by inductively coupled plasma spectrometry (ICP) on digested whole lagoon samples preserved with nitric acid. Microbial indicators were analyzed on whole lagoon samples using a commercial most probable number (MPN) method (IDEXX Laboratories, ME). Antibiotics were analyzed by the U.S. Geological Survey (Lawrence, KS) on filtered lagoon samples. Free estrogens and estrogen conjugates were analyzed on whole and filtered lagoon samples, respectively (Hutchins et al., 2007). More general information on nutrient characteristics and other properties of various types of lagoon water samples is also available in the literature (MWPS, 1993; NCSU, 1994; USDA, 1996; ASAE, 1999; USEPA, 2001).

Table 3. Antibiotic analyses of filtered lagoon samples from different CAFOs (mean with standard deviation, three locations for each lagoon).

CAFO† type	Tetracycline	Oxytetracycline	Chlorotetracycline	Iso-chlorotetracycline	Epi-iso-chlorotetracycline	Sulfamethazine	Lincomycin	Tylosin
	μg L^{-1}							
Beef feedlot‡	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.05 ± 0.08
Dairy‡	0.13 ± 0.07	<0.01	<0.01	0.01 ± 0.01	0.05 ± 0.08	<0.01	0.01 ± 0.00	<0.01
Poultry§	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.01 ± 0.02
Poultry‡	<0.01	<0.01	<0.01	0.02 ± 0.02	0.01 ± 0.01	<0.01	<0.01	<0.01
Poultry¶	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.01 ± 0.01	<0.01
Swine sow§	0.84 ± 1.45	<0.01	0.54 ± 0.47	26.7 ± 3.2	19.7 ± 2.6	<0.01	1.73 ± 1.52	<0.01
Swine finisher§	6.61 ± 6.50	0.14 ± 0.24	7.51 ± 6.73	97.3 ± 16.8	53.3 ± 5.1	<0.01	1340 ± 480	0.33 ± 0.30
Swine nursery§	<0.01	68.0 ± 15.4	<0.01	53.3 ± 9.3	22.3 ± 4.5	2.36 ± 1.22	38.0 ± 8.5	<0.01

† CAFO, concentrated animal feeding operation.

‡ Secondary lagoon.

§ Primary lagoon.

¶ Tertiary lagoon.

Nutrients and Organics

Table 1 provides illustrative concentrations of nutrients (NH_4–N, TKN, TP, and K) and organics (TOC) that were measured in several swine, poultry, dairy, and beef lagoon water samples. Ayers and Westcot (1989) reported severe restrictions on the use of irrigation water that had total N values greater than 30 mg L^{-1}. These same authors also reported that typical ranges for NH_4–N, PO_4–P, and K^+ in irrigation water was 0 to 5, 0 to 2, and 0 to 2 mg L^{-1}, respectively. The USEPA has set the maximum contaminant level (MCL) in ground water for nitrate (NO_3^-) and nitrite (NO_2^-) at 10 mg L^{-1} NO_3–N and 1 mg L^{-1} NO_2–N, respectively. The USEPA recommended guideline for TOC in drinking water is 4 mg L^{-1}. Hence, Table 1 indicates that nutrient and organic concentrations are very high in these lagoon water samples relative to irrigation and drinking water standards.

Published literature also indicates that animal wastes frequently contain high concentrations of nutrients (Chang and Entz, 1996; Hao and Chang, 2003) and organics rich in biochemical oxygen-demanding material (Webb and Archer, 1994) that can adversely impact soil and water quality (Jokela, 1992; Chang and Entz, 1996; Craun and Calderon, 1996; USEPA, 1997; USEPA, 2000). Potential environmental problems from excess amounts of nutrients and organics in water include algal blooms, reduced biodiversity, objectionable tastes and odors, and growth of toxic organisms in surface waters that are used for recreation and sources of drinking water (Mallin, 2000; Burkholder et al., 2007). These degraded conditions, especially the associated hypoxia/anoxia and high ammonia, have caused major kills of freshwater species (Burkholder et al., 2007). High nitrate levels in water have been associated with increased risk of methemoglobinemia for infants (blue-baby syndrome), as well as diarrhea and respiratory disease (Ward et al., 2005).

Heavy Metals

Excess amounts of metals are sometimes added to animal feeds to promote growth. For example, arsenic is often fed to chickens as organoarsenic compounds (e.g., roxarsone) to promote growth, kill parasites that cause diarrhea, and improve pigmentation of chicken meat (Hileman, 2007; Schaefer, 2007); copper and zinc are typically used in swine diets to promote growth (Parker et al., 1999). Table 1 provides illustrative concentrations of copper and zinc that were measured in several swine, poultry, dairy, and beef lagoon water samples. Ayers and Westcot (1989) reported that the maximum recommended concentration of arsenic, copper, and zinc in irrigation water is 0.10, 0.20, and 2.0 mg L^{-1}, respectively. Some of the lagoon water samples significantly exceeded the recommended copper and zinc concentrations for irrigation water, especially the samples from swine lagoon water. Possible adverse effects of high levels of these metals in the environment include increased risk of phytotoxicity, and surface and ground water contamination (McBride, 1995).

Table 4. Estrogen analyses of whole (free estrogen) and filtered (estrogen conjugate) lagoon samples from different CAFOs (mean with standard deviation, three locations for each lagoon).

CAFO† type	Estrone	17-α-estradiol	17-β-estradiol	Estriol	E1–3S‡	E2α-3S	E2β-3S	E2β-17S
	ng L^{-1}							
Beef feedlot§	17 ± 1	6 ± 1	<20	<8	<1	<1	<1	<1
Dairy§	76 ± 12	229 ± 56	153 ± 34	<8	87 ± 4	166 ± 22	42 ± 3	<1
Poultry¶	2970 ± 150	408 ± 37	64 ± 9	489 ± 49	1 ± 1	<1	<1	<1
Poultry§	1570 ± 80	131 ± 15	21 ± 10	190 ± 5	3 ± 1	<1	10 ± 3	<1
Poultry#	21 ± 2	<4	<20	<8	<1	<1	<1	<1
Swine sow¶	10500 ± 1260	1220 ± 70	211 ± 128	6290 ± 850	2 ± 0	<1	<1	80 ± 7
Swine finisher¶	1640 ± 10	184 ± 24	152 ± 44	1540 ± 30	<1	<1	<1	<1
Swine nursery¶	834 ± 73	74 ± 3	46 ± 32	353 ± 478	<1	<1	<1	<1

† CAFO, concentrated animal feeding operation.

‡ E1–3S denotes estrone-3-sulfate; E2α-3S denotes 17α-estradiol-3-sulfate; E2β-3S denotes 17β-estradiol-3-sulfate; E2β-17S denotes 17β-estradiol-17-sulfate.

§ Secondary lagoon.

¶ Primary lagoon.

Tertiary lagoon.

Salts

Table 1 provides illustrative concentrations of EC that were measured in several swine, poultry, dairy, and beef lagoon water samples. Ayers and Westcot (1989) reported severe restrictions on the use of irrigation water that had EC values greater than 3 dS m^{-1}. The salinity levels that are associated with these lagoon waters are very high relative to the irrigation quality guidelines.

Prolonged exposure of agricultural lands to animal wastes that have high salinity levels may alter many soil physical and chemical properties, and crop yields (Burns et al., 1985; King et al., 1985; Chang et al., 1991). For example, Chang et al. (1991) studied the effects of 11 yr of cattle manure addition on soil chemical properties. Accumulation of specific ions (soluble sodium, calcium, magnesium, chloride, sulfate, bicarbonate, and zinc), nutrients (total N, nitrate, total P, available P), and organic matter increased with increasing application rates of manure and lagoon water. Near the soil surface the EC and the sodium adsorption ratio (SAR) increased and the pH decreased with increasing addition rates of manure. Changes in these soil chemical properties can in turn influence soil hydraulic properties. For example, high SAR values are frequently associated with decreases in the saturated hydraulic conductivity (Ayers and Westcot, 1989) as a result of dispersion of the clay (colloidal) fraction of soils. High levels of EC as well as specific ions (sodium, chloride, boron, nitrate, and bicarbonate) can also severely influence crop yields (Maas and Hoffman, 1977; Maas, 1984) as a result of increases in osmotic stress and specific ion toxicities.

Pathogens

Animal wastes frequently contain pathogenic viruses, bacteria, and protozoa that pose a risk to human and/or animal health (USDA, 1992; USEPA, 1998; Gerba and Smith, 2005). Although more than 130 microbial pathogens have been identified from all animal species that may be transmitted to humans by various routes (USDA, 1992; USEPA, 1998), the most significant manure-borne zoonotic pathogens are the protozoan parasites *Cryptosporidium parvum* and *Giardia duodenalis*, and the bacterial pathogens *Salmonella* spp., *Campylobacter* spp., *Escherichia coli* O157:H7, and *Listeria monocytogenes*. Viruses of potential concern include: poliovirus, coxsackie virus, echovirus, hepatitis A, rotavirus, and Norwalk virus (Gerba and Smith, 2005).

The USEPA has set drinking water goals for pathogens to be no detection, because of the low infectious dose for many pathogens (Loge et al., 2002) and variability of individual responses to infection (Gerba, 1996). Regulations to protect public health from pathogens, however, are largely based on measured concentrations of indicator microorganisms such as total or fecal (thermo-tolerant) coliform (FC). For example, the World Health Organization (WHO) recommended standard for use of degraded water for irrigating crops eaten raw is <1000 FC per 100 mL (WHO, 2006), whereas in the United States the standard for unrestricted urban use varies with the state from nondetectable to 200 FC per 100 mL (USEPA, 2004). Table 2 provides illustrative concentrations of various indicator microorganisms of fecal contaminant (total coliforms, fecal coliforms, and fecal enterococci) that were measured in several swine, poultry, dairy, and beef lagoon water samples. The concentration of indicator microorganisms in the various lagoon water samples is very high, and significantly exceeds the recommended U.S. standards for unrestricted irrigation (USEPA, 2004).

Surface and ground water contamination by pathogenic microorganisms is common in many areas of the United States (USEPA, 1997). Drinking water exposures may occur in vulnerable private wells, whereas recreation exposures and illnesses can happen due to accidental ingestion of contaminated water and dermal contact during swimming. Surveys of waterborne disease outbreaks frequently demonstrate a farm animal source (Centers for Disease Control and Prevention, 1998). Pathogenic microorganisms in ground water have been estimated to cause between 750,000 and 5 million illnesses per year in the United States (Macler and Merkle, 2000). Greater risks of serious illness occur for the very young, elderly, pregnant women, the immunocompromised, and those predisposed with other illnesses (Gerba, 1996).

Manure-contaminated water resources have also been implicated in food-borne disease outbreaks on a variety of fresh produce (Gerba and Smith, 2005). The health impacts of such outbreaks can be very significant. For example, the 2006 outbreak of *E.coli* O157:H7 on spinach (*Spinacea oleracea* L.) from the Salinas Valley, California, resulted in 205 illnesses and 3 deaths (USFDA, 2007). Furthermore, loss of confidence in the safety of agricultural produce can have significant economic impacts. For example, the 2005 spinach crop from this same region was estimated to be worth around $188 million.

Antibiotics

Veterinary pharmaceuticals are routinely used in CAFOs as a therapeutic, for growth-improvement, and health-protection purposes. Antibiotics are a major group of veterinary pharmaceuticals (Boxall et al., 2003). In veterinary medicines, more than 70% of all consumed pharmaceuticals are antibiotic agents (Halling-Sorensen et al., 1998). It was estimated that livestock use of antibiotics in the United States was 9.8 million kg in 2001 (AHI, 2002). Two families of classic veterinary antibiotics, which are most heavily used, are tetracyclines (including chlorotetracycline, oxytetracycline, doxycycline, and tetracycline) and sulfonamides (including sulfanilamide, sulfadiazine, sulfadimidine, sulfadimethoxine, sulfapyridine, and sulfamethoxazole) (Thiele-Bruhn, 2003). Most of the antibiotics are not metabolized completely and are excreted from the treated animal shortly after medication. It was found that as much as 80% of the administered antibiotics occurred as parent compounds in animal wastes (Shore et al., 1988; Penprase, 2001).

Antibiotics possess a potential to adversely impact the environment (Halling-Sorensen, 2000; Burnison et al., 2003; Delepee et al., 2004; Fernandez et al., 2004; Halling-Sorensen et al., 2003a; Sengelov et al., 2003; Soto et al., 2004; Wollenberger et al., 2000). Little is currently known about the toxicity of antibiotic metabolites or degradates (Boxall et al., 2004), the potential synergistic effects of various mixtures of contaminants on target organisms (Sumpter and Johnson, 2005), and effects of the long-term exposure to low levels of antibiotics on environmental health (Daughton and Ternes, 1999). The greatest risks appear to be related to antibiotic resistance (Khachatourians, 1998; Kummerer, 2004) and natural ecosystem functions (Costanzo et al., 2005; Thiele-Bruhn

and Beck, 2005). In particular, increased antibiotic resistance of naturally occurring pathogens in manure-contaminated environments is of concern (Burkholder et al., 2007).

A recent USGS survey of 139 streams across 30 states in the United States found that 48% of these samples contained detectable levels of antibiotics (Kolpin et al., 2002). Although the origin of these antibiotics was not identified, this survey does indicate that antibiotics have been widely disseminated in the environment. Detection of veterinary antibiotics and/or antibiotic resistant bacteria in streams and aquifers near animal production operations and manure application sites has been reported (Hirsch et al., 1999; Chee-Sanford et al., 2001; Campagnolo et al., 2002; Brown et al., 2006; Sapkota et al., 2007; Gilchrist et al., 2007). Table 3 provides illustrative concentrations of several antibiotics that were measured in swine, poultry, dairy, and beef lagoon water samples. Concentrations ranged from less than 0.01 to 1340 $\mu g\ L^{-1}$ depending on the antibiotic and the lagoon source. At present antibiotics are not regulated or monitored by maximum contaminant levels in the Safe Drinking Water Act. The development of antibiotic resistance, however, has been reported to occur at very low antibiotic concentrations that are found in dilute soil and water environments (Baquero et al., 1998). Furthermore, Chee-Sanford et al. (2001) found on a swine CAFO that tetracycline resistance genes were identified in the swine, in the lagoon water, and in ground water that was 250 m downstream from the lagoon. Gilchrist et al. (2007) provide a review that gives further evidence that antibiotic resistance can develop at CAFOs.

Hormones

Animals eliminate estrogen, androgen, and gestagen hormones from their bodies in their feces and urine (Lange et al., 2002). The principal steroid hormones of general concern include 17α- and 17β-estradiol (estrogen), testosterone, 17α- and 17β-trenbolone (androgen), and progesterone and melengestrol acetate (gestagen), which are either excreted endogenously from livestock (natural hormones) or are produced as pharmaceuticals in veterinary clinical practices (exogenous chemicals). Regarding the synthetic hormones, both trenbolone acetate and melengestrol acetate are extensively used as growth promoters for cattle in the United States (Schiffer et al., 2001; Renner, 2002). Lange et al. (2002) estimated the amount of estrogens, androgens, and gestagens that cattle (45 Mg estrogens yr^{-1}, 1.9 Mg androgens yr^{-1}, and 253 Mg gestagens yr^{-1}), pigs (0.8 Mg estrogens yr^{-1}, 0.35 Mg androgens yr^{-1}, and 22 Mg gestagens yr^{-1}), and chickens (2.7 Mg estrogens yr^{-1} and 2.1 Mg androgens yr^{-1}) produced in the United States.

Table 4 provides illustrative concentrations of several estrogen hormones and associated conjugates that were measured in the swine, poultry, dairy, and beef lagoon water samples. Hormone concentrations ranged from <1 to 10,500 $ng\ L^{-1}$ depending on the estrogen and the lagoon source. Hanselman et al. (2003) has provided some additional information on the physical parameters for the various estrogenic hormones as well as their typical concentration ranges in animal wastes. Lange et al. (2002) provides information regarding typical concentration ranges for testosterone and progesterone in animal waste.

At present hormones are not regulated or monitored by maximum contaminant levels in the Safe Drinking Water Act. Steroid hormones, however, have been classified as highly potent endocrine-disrupting chemicals (EDCs), which may interfere with the normal function of the endocrine system of humans and animals (Ashby et al., 1997). Hanselman et al. (2003) reported that endogenous steroid estrogens (natural hormones) possess estrogenic potency 10,000 to 100,000 times higher than exogenous EDCs (excluding the synthetic estrogen used in birth control pills, ethinyl estradiol). Physiological and reproductive disorders in birds, fish, shellfish, turtles, gastropods, and mammals could be caused by EDCs (Colborn et al., 1993), including steroid hormones. Steroid hormones are a particular concern because there is evidence that very low concentrations of these chemicals can adversely affect the reproduction of fish and other aquatic species (Jobling et al., 1996, 1998; Hanselman et al., 2003; Ankley et al., 2003; Jensen et al., 2006). For example, when male fathead minnows (*Pimephales promelas*) were exposed to 0.12 $\mu g\ L^{-1}$ of 17β-estradiol, production of vitellogin (normally produced by females) was induced (Panter et al., 2000). Ankley et al. (2003) found that female fathead minnows exposed to 0.03 $\mu g\ L^{-1}$ of trenbolone acetate for 21 d developed tubercles, small bumps on the head normally found only in breeding males.

Kolpin et al. (2002) reported detection of steroid hormones in approximately 90% of the 139 sampled streams in a survey conducted across the United States. The source of these hormones was not identified in this study, but these findings suggest that hormones have been widely spread in the environment and there is concern that these EDCs may put ecosystems at risk. In addition to municipal wastewater treatment plants, CAFOs have been considered as an important source for the release of hormones into the environment. In contrast to sewage treatment plants that degrade estrogens fairly rapidly, CAFO lagoons typically function as holding reservoirs or anaerobic reactors, and waste effluent generally receives no additional treatment before land application. It has been estimated that estrogen loads from land application by livestock manure account for greater than 90% of the total estrogen in the environment (Khanal et al., 2006). Results presented in Table 4 indicate that high concentrations of estrogens in CAFO lagoon water could significantly elevate their concentrations in receiving surface waters and ground water. Numerous reports show that steroid hormones released from animal waste have been measured in both surface water and ground water (Shore et al., 1995; Nichols et al., 1997; Peterson et al., 2000; Finlay-Moore et al., 2000; Renner, 2002; Hanselman et al., 2003; Kolodziej et al., 2004).

Land Application

The USEPA currently requires that CAFO waste application to agricultural lands follow approved Nutrient Management Plans (NMPs). The National Resources Conservation Service (NRCS) defines a NMP as, "Managing the amount, source, placement, form and timing of the application of nutrients and soil amendments" (USDA, 2000). The purpose of a NMP is to meet the nutrient needs of the crop to be grown, while minimizing the loss of nutrients to surface and ground water (USEPA, 2003). Nutrient management plans are developed to adhere to state-specific NRCS

Table 5. Nutrient uptake parameters for selected crops (Kellogg et al., 2000).

Crop	Nitrogen	Phosphorus
	kg per Mg of product	
Corn for silage	3.6	0.5
Sorghum for silage	7.4	1.2
Barley	18.6	3.8
Wheat	20.6	3.6
Alfalfa	25.2	2.4
Grass silage	6.8	0.8

guidelines. In general, a NMP is developed by considering all nutrient input sources (such as manure, fertilizer, lagoon water, and well water), the nutrient content at the soil surface, nutrient volatilization losses to the atmosphere (e.g., nitrogen losses as ammonia), nutrient mineralization rates, and plant uptake rates for nutrients. The nutrient concentrations are typically measured directly for the input sources, at the soil surface, and in plant tissues, whereas nutrient volatilization and mineralization rates are commonly estimated from literature values. The amount of lagoon water or other nutrient source that can be used in a given irrigation cycle is determined based on nutrient mass-balance considerations for a limiting nutrient (e.g., N or P). A review of several state NMP guidelines shows that they are typically written so as to be protective of surface water resources, and only provide limited direction on land application site characterization and design to protect ground water from contamination (such as in regions with shallow water tables).

A brief example of NMP implementation is given below. In this example we assume that N is the limiting nutrient for plant growth, and use the CAFO lagoon water composition information for nutrients presented in Table 1. Table 5 lists nutrient uptake parameters for selected crops (Kellogg et al., 2000) that were used in this example. For simplicity we also make the following assumptions: N volatilization loss of 20%, N mineralization (ammonification) rate of 50%, initially zero N in the soil profile and in the well water, and the only water source to the plants was irrigation water. Table 6 presents the blending ratios that were calculated to meet the nutrient (N) requirements for winter wheat (*Triticum aestivum* L.) and summer corn (*Zea mays* L.) for silage assuming final yields of 27 and 56 Mg ha^{-1}, respectively, and 90 d of full cover crop. We performed this calculation for two climatic conditions, inducing different evapotranspiration (ET) fluxes that correspond to semiarid and temperate areas. Table 6 indicates that the higher ET flux produces a higher blending ratio. Lagoon water with a low N concentration (dairy and beef) provides insufficient N at low ET fluxes and therefore an additional N source is required (fertilizer) to meet crop needs.

Data in Tables 1 to 6 can be used to provide an illustrative estimate of the environmental loading for the various contaminants identified in the lagoon water under NMP application conditions. As an example, Table 7 provides the estimated environmental loading of total salts (from EC values given in Table 1), metals (sum of Zn and Cu in a given row of Table 1), indicator microorganisms (sum of total coliform and enterococcus in a given row of Table 2), antibiotics (sum of all antibiotics in a given row of Table 3), and estrogen hormones (sum of all hormones in a given row of Table 4) when the various lagoon waters (blended with well water) were applied to a 1-m^2 area of agricultural field to meet the N needs of corn during a 90-d summer growing season with an evapotranspiration rate of 10 mm d^{-1}. In contrast to Tables 1 through 4, Table 7 is dependent on both the contaminant concentration in the lagoon water and on the blending ratio given in Table 6.

Nutrient management plans assume that a limiting nutrient is the primary environmental concern. To minimize surface water runoff and leaching to ground water, NMPs also imply that water is applied to the crop to meet the potential evapotranspiration demands. The environmental loading information presented in Table 7 indicates that significant amounts of salts, heavy metals, indicator microorganisms, antibiotics, and hormones will be added to agricultural lands where a NMP is being implemented. An implicit assumption of a NMP is that all CAFO contaminants will be taken up, inactivated, retained, or degraded in the root zone, so that surface and ground water is inherently protected. The validity of these assumptions for all lagoon water contaminants has not yet been thoroughly studied. Below we discuss possible weaknesses in NMP design and application, as well as potential transport pathways and processes for various lagoon water contaminants.

Table 6. Hypothetical blending ratio (well water to lagoon water) for various lagoon waters when winter wheat and summer corn are grown using semiarid and temperate climates.

		Winter wheat blending ratio		Summer corn blending ratio	
CAFO† type	Plant available N‡	Semiarid climate ET = 5 mm d^{-1}	Temperate climate ET = 2.5 mm d^{-1}	Semiarid climate ET = 10 mm d^{-1}	Temperate climate ET = 5 mm d^{-1}
	mg L^{-1}				
Beef feedlot§	41	insufficient N	insufficient N	0.93	0.46
Dairy§	118	0.05	insufficient N	4.56	2.28
Poultry¶	598	3.88	1.43	27.46	12.90
Poultry§	290	1.37	0.18	12.79	5.74
Poultry#	65	insufficient N	insufficient N	2.07	0.50
Swine sow¶	928	6.58	2.78	43.22	20.59
Swine finisher¶	1704	12.93	5.94	80.21	38.66
Swine nursery¶	1431	10.70	4.82	67.19	32.30

† CAFO, concentrated animal feeding operation. ET, evapotranspiration.

‡ Total N minus amount due to volatilization and ammonification.

§ Secondary lagoon.

¶ Primary lagoon.

Tertiary lagoon.

Table 7. The estimated environmental loading of total salts, metals, antibiotics, estrogen hormones, and indicator microorganisms when the various lagoon waters were applied to a 1 m^2 area of agricultural field to meet the nitrogen needs of corn during a 90 d summer growing season. An evapotranspiration rate of 10 mm d^{-1} was assumed.

CAFO† type	Total salts	Total metals (Zn + Cu)	Total estrogen hormones	Total antibiotics	Total indicator microbes
	g m^{-2}	mg m^{-2}	μg m^{-2}		no. m^{-2}
Beef feedlot‡	1755.0	387.00	49.50	108.00	6.1E+07
Dairy‡	458.9	213.16	150.39	45.39	3.8E+10
Poultry§	277.8	78.66	128.97	2.62	6.2E+07
Poultry‡	422.2	22.52	122.23	4.93	6.2E+07
Poultry¶	1434.8	39.13	24.78	34.78	2.7E+08
Swine sow§	196.8	324.43	250.20	475.61	1.8E+08
Swine finisher§	163.3	194.90	22.22	15,797.41	2.1E+08
Swine nursery§	216.0	1638.19	12.83	1,750.57	5.8E+07

† CAFO, concentration animal feeding operation.

‡ Secondary lagoon.

§ Primary lagoon.

¶ Tertiary lagoon.

Water Flow

Figure 1 provides a schematic of the root zone at a NMP application site (Simunek and van Genuchten, 2007). The basic premise of a NMP is that water and nutrients are applied to the root zone at agronomic rates so that both surface and ground water resources will be protected from all contaminants in lagoon water. Potential water inputs at a NMP application site include irrigation water (well and/or CAFO lagoon water), precipitation, surface water, snowmelt, soil water, and shallow ground water tables. The water application amount per irrigation should be based on measurements or estimates of potential evapotranspiration (PET) and water-balance considerations at the application site. If water input at a NMP application site equals PET, then surface and ground water contamination should theoretically be eliminated because all of the applied water to the site is used by the crops.

When water in excess of PET is applied to a NMP site, then both surface and ground water contamination problems are possible. This situation can arise from a number of different scenarios that may be beyond human control. Potential problems may arise due to inaccuracies in the estimation of PET, or from not accounting for all the sources of plant-available water (perched water tables, precipitation, snowmelt, and surface water runoff). For example, significant amounts of unexpected precipitation (a thunderstorm) after application of nutrients may lead to surface water runoff and/or leaching of contaminants. Other problems can occur due to nonuniform water application practices during flood or furrow irrigation or as a result of spatial variability of soil hydraulic properties (e.g., different amounts of water are applied to specific locations in the field) and evapotranspiration. Changes in surface topography that produce surface water runoff and lateral flow to other locations in the field pose other NMP difficulties.

Nutrient management plans implicitly assume that water flow and lagoon water contaminant transport are controlled by the soil matrix. Preferential flow is a potentially important mechanism for water and contaminant transport to bypass portions of the soil matrix and root zone. Several recent reviews and other papers provide detailed discussions of the various processes and conditions leading to preferential flow (Ritsema et al., 1993; de Rooij, 2000; Evans et al., 2001; National Research Council, 2001; Bodvarsson et al., 2003; Simunek et al., 2003; Wang et al., 2004). In summary, preferential water flow can occur as a result of funneling of water at textural interfaces (capillary barriers), unstable flow behavior that is induced by spatial and/or temporal variations in capillary characteristics (wettability or hysteresis), dynamic capillary properties (nonequilibrium capillary pressure- water content characteristics), macropores, and fractured systems. All of these factors can potentially accelerate the movement of lagoon water contaminants through the root zone to underlying ground water. Some mechanisms of preferential flow are also anticipated to depend on the initial soil water content and water application rate. For example, soil water repellency is reported to occur below some critical water content (Ritsema and Dekker, 2000), hysteresis is induced when

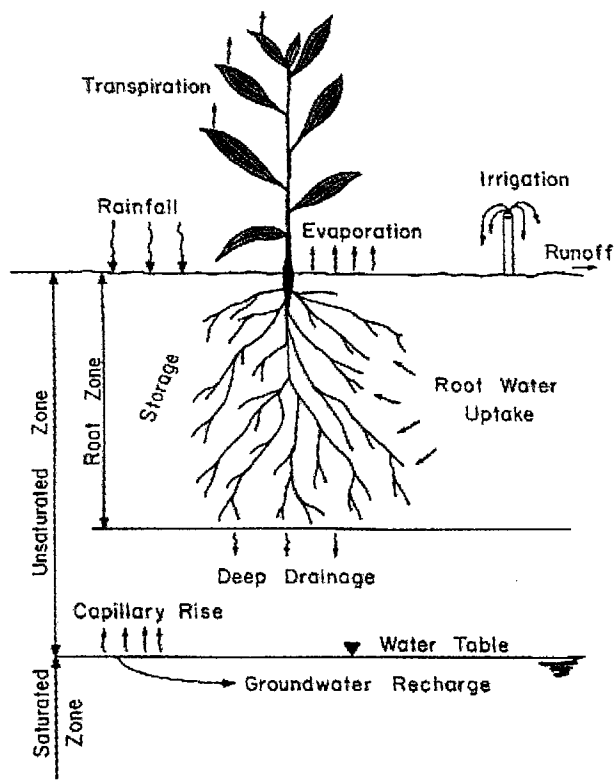

Fig. 1. A schematic of the root zone at a nutrient management plan application site (from Simunek and van Genuchten, 2007).

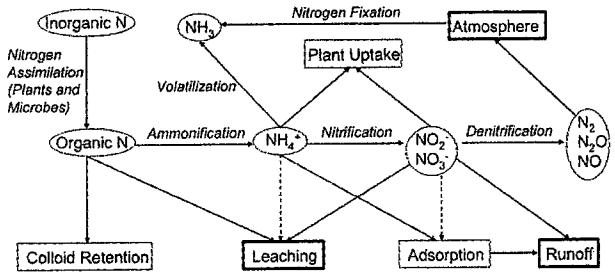

Fig. 2. A schematic of the processes and N species that are involved in the N cycle. Important inorganic forms of N are ammonium ions (NH_4^+), ammonia gas (NH_3), nitrite (NO_2^-), nitrate (NO_3^-), nitrogen gas (N_2), nitrous oxide (N_2O), and nitric oxide (NO). The dashed lines denote secondary transport processes.

infiltration stops and redistribution occurs (Wang et al., 2003), and filling of macropores occurs primarily near saturated conditions (Mohanty et al., 1997). Improved NMP designs are needed to provide guidelines to minimize transport processes that bypass the root zone. For example, it may be possible to minimize preferential flow in macropores by applying irrigation water at rates lower than the saturated conductivity of the soil.

Nutrient Transport

It should be mentioned that NMPs are only as good as the information on evapotranspiration and nutrient mass balance. Errors in water and nutrient application rates, or nonuniform water application, will likely lead to excesses and/or shortages in lagoon water application in certain locations. Such errors are potentially responsible for some of the reported environmental problems associated with lagoon water application sites (Evans et al., 1984; King et al., 1985; Sims et al., 1998; Correll, 1998). Another potential problem arises when the relative content of N and P in lagoon water differs from that in the crops. In this case, NMPs that are designed to meet the N requirement for crops may result in the over-application of P. Conversely, NMPs based on crop P needs may significantly reduce the lagoon water application rates, and thus require N fertilizers to meet plant needs.

The biogeochemistry of nutrients at NMP sites can be quite complex. For example, Fig. 2 shows a schematic of the processes and N species that are involved in the N cycle. The most important inorganic forms of N are ammonium ions (NH_4^+), ammonia gas (NH_3), nitrite (NO_2^-), nitrate (NO_3^-), nitrogen gas (N_2), nitrous oxide (N_2O), and nitric oxide (NO). Organic nitrogen compounds in lagoon water occur as urea, amino acids, amines, purines, and pyrimidines, as well as a fraction of the dry weight of plants, microorganisms, detritus, and soils. Organic and inorganic forms of N are continually involved in biogeochemical transformations. The major processes involved in the N cycle are ammonification (mineralization of organic N to NH_4^+), nitrification (oxidation of NH_3 to NO_3^-), denitrification (reduction of NO_3^- to N_2), N fixation (reduction of N_2 to NH_3), N assimilation (conversion of inorganic N to organic forms), and N volatilization (conversion of NH_4^+ to NH_3).

The biogeochemistry of P at NMP sites is also very complex. Most P (63–92%) in manure and lagoon water occurs in inorganic forms (Sharpley and Moyer, 2000). Bohn et al. (1985) presents a speciation diagram of P species of dissolved orthophosphoric acid (H_3PO_4) as a function of pH. The three acid dissociation constants for H_3PO_4 at 25°C are 7.5×10^{-3} ($pK_{a1} = 2.1$), 6.2×10^{-8} ($pK_{a2} = 7.2$), and 5.0×10^{-13} ($pK_{a3} = 12.3$). The dominant phosphate species in the pH range of 2.1 to 7.2 is $H_2PO_4^-$, whereas HPO_4^{-2} is the main species in the pH range of 7.2 to 12.3. Both of these species are available for plant uptake. Phosphorus, however, forms complex minerals with many elements (Arai and Sparks, 2007). For example, P species frequently precipitate out of solution with Ca, Fe, and Al ions (Yeoman et al., 1988; Moore and Reddy, 1994). Furthermore, P species can strongly adsorb to positively charged surfaces in soils (Torkzaban et al., 2006b; Arai and Sparks, 2007), such as on metal oxide surfaces and clay edges. The solubility of P in soil solution is, therefore, a complex function of biogeochemical variables and processes such as pH, oxidation–reduction potential, the concentrations of cations (such as Ca^{2+}, Mg^{2+}, Al^{3+}, and Fe^{3+}) and P species, soil mineralogy and organic matter content, and the mineralization rate of organic (manure) compounds (Moore and Reddy, 1994; Sharpley, 1995; Arai and Sparks, 2007). Measurements of total P in soils may, therefore, not accurately assess the potential for plant uptake of P and the potential for subsurface transport losses (Brock et al., 2007).

The subsurface fate of nutrient species at NMP sites depends on a variety of transport and mass transfer processes. For example, dissolved inorganic N ions such as NH_4^+, NO_2^-, and NO_3^- can be transported with flowing water (advection), as well as by diffusive and dispersive solute fluxes. Interface mass transfer processes may play important roles between dissolved N species and soil–sediment surfaces (e.g., sorption–desorption of NH_4^+) or the air phase (e.g., volatilization of NH_3), and uptake of N species by plants and microorganisms is anticipated. The presence of colloidal forms (manure suspension) containing organic N and P that are mobile in the water phase provide another nutrient transport pathway.

Environmental losses of nutrient species that are primarily associated with the solid phase (such as NH_4^+ and P) will occur mainly in conjunction with high sediment loads during surface water runoff (Sims et al., 1998; Correll, 1998). For a well-designed NMP that is conducted on most mineral soils, little leaching and subsurface transport of P is expected. Sims et al. (1998), however, review the literature that indicates that P species may also be leached through soils under certain environmental conditions (e.g., deep sandy soils, high organic matter soils, or soils with high soil P concentrations from long-term overfertilization and/or excessive use of organic wastes). Pautler and Sims (2000) suggested that the degree of P saturation, DPS (defined as the ratio of the amount of P sorbed by the soil to the P sorption capacity), is a better indicator for the potential losses to subsurface transport than the total P. Increased addition of manure has been reported to nonlinearly increase the P sorption capacity of the soil (Brock et al., 2007). When the DPS was greater than around 30%, P transport from soils increased dramatically (Brock et al., 2007).

Pathogen Transport

The root and vadose zones at NMP application sites play a critical role in protecting water supplies from pathogen contamination. Effective treatment relies on the retention and inactiva-

tion of pathogens in unsaturated or variably saturated porous media. Inactivation of pathogenic microorganisms is commonly assumed to occur within 60 d, although many viruses and protozoa are known to be viable for a longer duration (Schijven et al., 2006). Considerable research has been devoted to the fate and transport of microorganisms and other colloids in porous media (reviews are given by Herzig et al., 1970; McDowell-Boyer et al., 1986; McCarthy and Zachara, 1989, Ryan and Elimelech, 1996; Khilar and Fogler, 1998; Schijven and Hassanizadeh, 2000; Harvey and Harms, 2002; Jin and Flury, 2002; Ginn et al., 2002; de Jonge et al., 2004; DeNovio et al., 2004; Rockhold et al., 2004; Sen and Khilar, 2006; Tufenkji et al., 2006). Most of the published research pertains to saturated media, less is known about microbe transport and retention in unsaturated systems (Wan and Wilson, 1994b; Choi and Corapcioglu, 1997; Wan and Tokunaga, 1997; Schafer et al., 1998a, 1998b; Saiers et al., 2003; Saiers and Lenhart, 2003; de Jonge et al., 2004; DeNovio et al., 2004). Below we briefly review the literature about processes and factors that will potentially influence the transport and fate of pathogens and surrogates (indicator microorganisms, latex microspheres, and other colloids) at NMP sites.

Microorganism retention mechanisms in the vadose zone are more complicated than in the saturated zone, mainly due to the presence of air in the system. In unsaturated porous media water flow is restricted by capillary forces to the smaller regions of the pore space and flow rates are relatively small. Microorganism transport may be influenced by increased attachment to the solid–water interface (Chu et al., 2001; Lance and Gerba, 1984; Torkzaban et al., 2006a), attachment to the air–water interface (Wan and Wilson, 1994a, 1994b; Schafer et al., 1998b; Torkzaban et al., 2006b), straining of microorganisms near multiple interfaces in the smallest regions of the pore space (McDowell-Boyer et al., 1986; Cushing and Lawler, 1998; Bradford et al., 2006a), film straining in water films enveloping the solid phase (Wan and Tokonaga, 1997; Saiers and Lenhart, 2003), and retention at the solid–air–water triple point (Crist et al., 2004, 2005; Chen and Flury, 2005; Zevi et al., 2005; Steenhuis et al., 2006). Transients in water content and composition during infiltration and drainage processes can also significantly influence these unsaturated retention mechanisms (Saiers et al., 2003; Saiers and Lenhart, 2003; Torkzaban et al., 2006b).

Size exclusion affects the mobility of microorganisms in the vadose zone by constraining them to more conductive flow domains and large pore networks that are physically accessible (Ryan and Elimelech, 1996; Ginn, 2002). As a result, microbes can be transported faster than a conservative solute tracer (Cumbie and McKay, 1999; Harter et al., 2000; Bradford et al., 2003). Differences in the dispersive flux for microbes and a conservative solute tracer may also occur as a result of size exclusion (Sinton et al., 2000; Bradford et al., 2002).

Various combinations of well, surface, and lagoon water, encompassing a range in solution chemistry, are used for irrigation at NMP sites. Many chemical factors (i.e., pH, ionic strength, surface charge, etc.) are known to influence the transport behavior of pathogens. For example, using colloids as pathogen models, colloids are stabilized when the electrical double layers are expanded and when the net particle charge does not equal zero (Ouyang et al., 1996). Increasing electrolyte concentration and ionic strength decreases the double layer thickness and thereby promotes aggregation and microorganism–porous medium interactions. Aggregation will in turn impact the colloid size distribution and therefore straining. Aqueous phase pH influences the net microorganism and solid surface charge by changing pH-dependent surface charge sites. McCarthy and Zachara (1989) reported that colloids can be produced through disaggregation when the ion balance is shifted from one dominated by Ca^{2+} to one dominated by Na^{+}. These chemical factors (pH, ionic strength, surface charge, and chemical composition) are also known to influence soil structure (disaggregation) and pore-size distribution (shrinking and swelling) when the soil contains clays and other colloidal materials (Ayers and Westcot, 1989).

Most transport experiments with pathogens have been conducted in the absence of dissolved manure suspensions. Pathogens in lagoon water, however, constitute only a small portion of the colloidal suspension. Large quantities of manure components are also present. Several researchers have examined the influence of organic matter or manure suspensions on microbe transport (Johnson and Logan, 1996; Pieper et al., 1997; Powelson and Mills, 2001; Guber et al., 2005a, 2005b; Bradford et al., 2006c, 2006d). Some researchers reported that dissolved organic matter (DOM) or manure suspensions enhanced microbe transport (Johnson and Logan, 1996; Pieper et al., 1997; Powelson and Mills, 2001; Guber et al., 2005a, 2005b; Bradford et al., 2006c, 2006d). Blocking of favorable attachment sites by organic matter has typically been used to explain this enhanced transport (Johnson and Logan, 1996; Pieper et al., 1997; Guber et al., 2005a, 2005b), although filling of straining sites provides an alternative explanation (Bradford et al., 2006c). Dissolved organic matter has also been reported to sorb onto bacterial cell walls and alter their electrophoretic mobility (Gerritson and Bradley, 1987). Increasing the negative charge of the bacterial surface diminishes its attachment onto negatively charged solid surfaces (Sharma et al., 1985). Other researchers have reported that organic matter inhibits microbe transport due to hydrophobic interactions between microbes and grain surfaces that are coated with organic matter (Bales et al., 1993). Adsorption of pathogens onto mobile manure colloids could also facilitate their transport potential (Jin et al., 2000; de Jonge et al., 2004).

As an illustration of the potential significance of manure suspension (surrogate for lagoon water) on pathogen transport at a NMP site, Fig. 3 presents breakthrough curves for pathogenic *E. coli* O157:H7 in various solutions without and with manure components (0.001 M NaBr buffered to a pH of 6.7 using 5×10^{-5} M $NaHCO_3$; and 4 g L^{-1} of dairy calf manure suspended in this same 0.001 M NaBr solution and then filtered through 103 μm stainless screen) that were passed through 150 μm Ottawa sand. Before discussing the results, several details on the experimental procedures are given below.

To minimize microbial growth and activity, these column experiments were conducted in a constant temperature room set at 4°C. The porosity of the columns was 0.32 cm^3 cm^{-3}, and the Darcy velocity was approximately 0.1 cm min^{-1}. *Escherichia coli* O157:H7/pGFP strain 72 (Dr. Pina Fratamico, USDA-

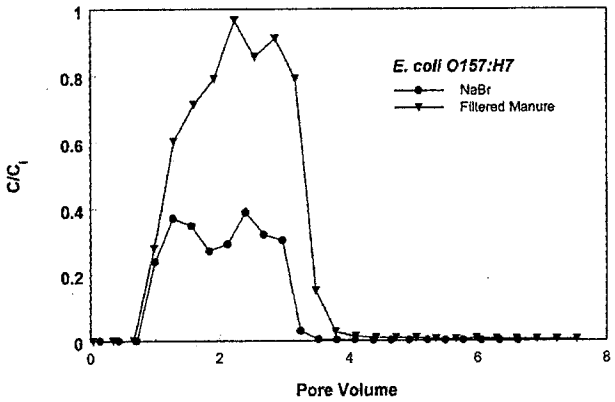

Fig. 3. A plot of the relative effluent concentration (C/C_i where C and C_i are the *E. coli* O157:H7 concentrations in the effluent and influent tracer solutions, respectively) as a function of pore volumes for pathogenic *E. coli* O157:H7 in various solutions without and with manure components (0.001 M NaBr buffered to a pH of 6.7 using 5×10^{-5} M $NaHCO_3$; and 4 g L^{-1} of dairy calf manure suspended in this same 0.001 M NaBr solution and then filtered through 103-μm stainless screen) that were passed through 150-μm Ottawa sand.

ARS-ERRC, Wyndmoor, PA) was grown in tryptic soy broth with ampicillin (50 g mL^{-1}) overnight at 37°C (Fratamico et al., 1997). The bacteria were washed twice with phosphate buffered saline solution, resuspended in the 0.001 M NaBr solution, and diluted to the desired concentrations (ranged from 0.46×10^5 to 1.7×10^5 cell mL^{-1}). The *E. coli* O157:H7 concentrations in the influent and effluent solution were quantified using the spread plating method (Clesceri et al., 1989) on tryptic soy agar plates with ampicillin (50 g mL^{-1} final concentration).

Figure 3 indicates that *E. coli* O157:H7 transport in the presence of manure suspension was much greater (82% in effluent) compared to in the absence (35% in effluent) of manure suspension. Transport differences for *E. coli* in the presence and absence of dissolved manure are likely induced by variations in the aqueous chemistry of the tracer solution. The hydrophobicity (contact angle measured with a Tantec Contact Angle Meter on a lawn of bacteria equaled 18°) and size (1.35–1.52 μm measured with a ZetaPals instrument) of clean and manure equilibrated *E. coli* were similar. The pH (6.7–7.6) and electrical conductivity (0.14–0.26 dS m^{-1}) of both 0.001 M NaBr solution and manure suspension were also comparable. In contrast, manure equilibrated *E. coli* was approximately four times more negatively charged than the clean *E. coli* (−25.4 mV compared with −6.8 mV measured with a ZetaPALs instrument), suggesting that sorption of dissolved manure compounds significantly altered the surface charge characteristics of these bacteria. It is also possible that manure particles filled straining locations, and therefore diminished the amount of bacteria that otherwise would be retained in these locations (Bradford et al., 2006b, 2006c).

Many microbiological factors will also play important roles in pathogen migration at NMP sites. Microbe growth, death, and inactivation are of special importance. The ability of many pathogens to survive in manure (Wang et al., 1996), lagoon water (Ibekwe et al., 2003), and soil (Gagliardi and Karns, 2000, 2002) has been well documented. The survival and subsequent transport of viruses have also been reported to be enhanced in the presence of manure suspensions (Bradford et al., 2006d), likely due to sorption of organic components onto metal oxide surfaces that would otherwise inactivate the viruses (Sagripanti et al., 1993; Pieper et al., 1997; Schijven et al., 1999; Chu et al., 2001; Ryan et al., 2002). Some microbes can also alter their surface characteristics and surrounding environment so as to promote attachment or release (detachment) (Ginn et al., 2002). Other mobile microbes possess the ability to move in the direction of increasing concentration gradient of chemoattractants or a decreasing concentration gradient of chemorepellents (Nelson and Ginn, 2001). Many of these microbiological processes are expected to be strongly coupled with temperature and nutrient conditions at NMP sites.

Hormones and Antibiotics

Application of animal wastes to agricultural land may serve as an important pathway to disseminate antibiotics and hormones in the environment (Thiele-Bruhn, 2003; De Liguoro et al., 2003). These chemicals need to be retained and/or degraded in the root zone at NMP sites to protect surface and ground water resources from contamination. Limited studies have been conducted on the environmental persistence, sorption, and transport of various pharmaceutical compounds (reviews are provided by Daughton and Ternes, 1999; Tolls, 2001; Renner, 2002; Hanselman et al., 2003; Thiele-Bruhn, 2003; Khanal et al., 2006). This information is briefly discussed below.

Sorption of hormones and antibiotics to soil surfaces is dependent on the chemistry of the compound, the aqueous phase, and the solid interface. Hormones and antibiotics commonly have polar functional groups that are ionizable within the pH range of natural soils (Jorgensen and Halling-Sorensen, 2000; Jones et al., 2005; ter Laak et al., 2006; Kahle and Stamm, 2007). These chemicals may occur as negative, neutral, zwitterionic, and positively charged species that have sorption behavior that is dependent on the solution pH and other environmental conditions (ter Laak et al., 2006; Kahle and Stamm, 2007). Many antibiotics and hormones are also lipophilic (hydrophobic) compounds that adsorb strongly to the organic fraction of soils (Tolls, 2001; Thiele-Bruhn, 2003).

Hormones and antibiotics may undergo a variety of abiotic and biotic transformations in the environment. Potentially important abiotic degradation processes in lagoon water include hydrolysis and photodegradation. Hydrolysis of antibiotics is dependent on the solution pH (Volmer and Hui, 1998). Several researchers have studied the potential for photodegradation of hormones and antibiotics in surface waters (Boreen et al., 2003; Andreozzi et al., 2003; Lin and Reinhard, 2005). Little research, however, has addressed the potential for photodegradation in lagoon water. Biodegradation of organic compounds occur in a wide variety of environments (Vidali, 2001). Many factors will influence the rate and extent of biodegradation including: structure and composition of the chemical, microbial community, available C sources, electronic acceptors, the nutrients, pH, temperature, oxidation–reductive potential, and water content (Vidali, 2001). Metabolism and degradation of antibiotics and hormones can transform parent compounds to more water-

soluble metabolites (Halling-Sorensen et al., 1998). Metabolites may, therefore, exhibit different transport potentials than parent compounds as a result of dissimilar degradation and sorption rates (Colucci et al., 2001; Colucci and Topp, 2001; Casey et al., 2003, 2004). Knowledge of the transport behavior for both parent and metabolites is desirable, as certain pharmaceutical degradation products may be of equal or greater health concern.

Steroid estrogens 17α-estradiol, 17β-estradiol, estrone, and estriol and androgen testosterone have been receiving extensive attention as the most widely present hormones in the environment. Recent batch (Lee et al., 2003) and column studies (Das et al., 2004; Casey et al., 2003, 2004, 2005) have found that these hormones can have relatively short half-lives and a high affinity for sorption in natural soils. Casey et al. (2003) applied 17β-estradiol to the top of the soil column and found traces of estriol and a highly polar metabolite (unidentified) in the effluent. Meanwhile, 17β-estradiol, estrone, and traces of estriol were sorbed to the soil within the column. However, definitive mechanisms of hormone sorption and transformation in soil are still not fully understood. It appears that estrogens are readily biodegraded in the soil, manure, and manure-containing wastewater. For example, 17β-estradiol may be readily oxidized to estrone in the environment (Lai et al., 2000; Colucci et al., 2001). A similar transformation has also been observed in lagoons containing wastes from swine (Fine et al., 2003). However, the transformation is reversible under anaerobic conditions, which may produce 17α-estradiol from estrone via racemization in addition to 17β-estradiol (Czajka and Londry, 2006). Of these estrogens, 17β-estradiol is the most potent EDC, whereas 17α-estradiol and estrone are identified as the less active estrogens. Estriol is considered to have the least potency as an EDC. Hence, processes of transformation and metabolism of hormones can alter their biological activity in the aquatic environment.

Generally, natural hormone excreted from livestock species are either free steroids or sulfate or glucuronide conjugates. A recent report shows that estrogen conjugates contributes significantly to the overall estrogen load in various CAFO lagoons (Hutchins et al., 2007). For example, these researchers estimated that the corresponding contributions of estrogen conjugates were 57% for the dairy lagoon, 95% for the poultry tertiary lagoon, and ranged from 27 to 35% for the swine nursery, beef feedlot, and poultry primary lagoons. Although most conjugated forms of the hormones are biologically inactive, they can act as precursor hormone reservoirs that may be readily deconjugated by bacteria to produce corresponding active free hormones (Hanselman et al., 2003). Hence, the effect of steroid conjugates on the total endocrine-disrupting activity of estrogens in the environment should not be ignored. Estrogen conjugates are also expected to be more polar and therefore more mobile in the soil.

In contrast to natural hormones, relatively little is known about the environmental fate of the synthetic steroid hormones (trenbolone acetate and melengestrol acetate) and their subsequent degradation products (α-trenbolone, β-trenbolone, and trendione). Schiffer et al. (2001) reported that melengestrol acetate and trenbolone can persist in the soil many months after land application. The degradation products α-trenbolone and β-trenbolone have been demonstrated to cause reproductive effects on fathead minnow (Ankley et al., 2003; Jensen et al., 2006).

As the most heavily used veterinary antibiotic family, tetracyclines have gained extensive attention. Tetracyclines are polyketides and comprised of a naphthalene ring structure. They are amphoteric compounds with three pK_a values at 3.3, 7.7, and 9.3 and are more stable in acidic than in basic media (Halling-Sorensen et al., 2002). Most tetracyclines are sparingly soluble in water, strongly adsorbed in soil, and persistent in the environment (Rabolle and Spliid, 2000; Thiele-Bruhn, 2003; De Liguoro et al., 2003). Tetracyclines were found to chelate with divalent cation and β-diketones and strongly bind to proteins and silanolic groups (Oka et al., 2000). Degradation pathways of oxytetracycline in a soil–water system have been investigated and several degradation products have been identified (Halling-Sorensen et al., 2003b; Loke et al., 2003).

As the second most frequently used group of veterinary antibiotics, sulfonamides have also been studied to understand their environmental behaviors. Sulfonamides are relatively insoluble in water but were found to be highly mobile in the soil (Thiele-Bruhn, 2003; Boxall et al., 2002). Limited information regarding the degradation kinetics and pathways of sulfonamides in the environment is available. Sulfadimethoxine has been reported to degrade fast in manure, with a half-life of 1.4 to 2.6 d depending on the initial concentration of sulfadimethoxine, manure moisture, and storage temperature (Wang et al., 2006a). Degradation of sulfadimethoxine in soil was found to be much slower compared with in manure, indicating that sulfadimethoxine became more persistent once it is released from contaminated manure into soil (Wang et al., 2006b). A photo-catalytic degradation study of several sulfonamides on TiO_2 implies a possible cleavage of the S–N bond during their degradation in the environment (Caza et al., 2004).

The transport potential of antibiotics and hormones can be enhanced when these compounds are strongly adsorbed to soil and/or manure colloids that are mobile in the aqueous phase (Tolls, 2001; Thiele-Bruhn, 2003; Hanselman et al., 2003; Holbrook et al., 2004; Zhou et al., 2007). The concentration of total suspended solids in lagoon water (Table 1) and dissolved manure in runoff water (Bradford and Schijven, 2002) can be very high. Hence, failure to account for colloid-facilitated transport may severely underestimate the transport potential of antibiotics and hormones in risk assessment. Colloid-facilitated transport has been illustrated in the literature for numerous contaminants, including heavy metals (Grolimund et al., 1996), radionuclides (Von Gunten et al., 1988; Kim et al., 1992; Noell et al., 1998), pesticides (Vinten et al., 1983; Kan and Tomson, 1990, Lindqvist and Enfield, 1992), antibiotics (Tolls, 2001; Thiele-Bruhn, 2003), hormones (Hanselman et al., 2003), and other contaminants (Magee et al., 1991; Mansfeldt et al., 2004). Several reviews on colloid facilitated transport of various contaminants are available in the literature (McCarthy and Zachara, 1989; de Jonge et al., 2004; Simunek et al., 2006).

Treatments

Figure 4 presents a flowchart that illustrates the potential steps that may be used to assess and improve the performance of lagoon water application sites. To determine whether the reuse of CAFO wastewater on agricultural lands poses an acceptable

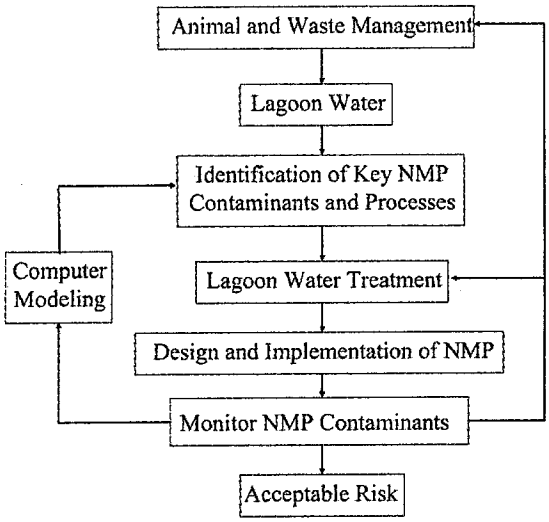

Fig. 4. A flowchart that illustrates the steps that may be used to assess and improve the performance of lagoon water application sites.

environmental risk, it is necessary to monitor the transport and fate of relevant contaminants at these sites. At present land application of CAFO lagoon water is based on a nutrient balance. As mentioned earlier, other potential contaminants of concern at NMP sites include salts, heavy metals, pathogens, antibiotics, and hormones. If the risk (measured concentration of a particular contaminant below or in the root zone) is deemed to be unacceptable, then management practices for animals will have to be modified and/or wastewater treatment will have to be implemented before this water can be used in NMPs. Based on current technologies and associated costs, it is not practical for this level of monitoring to be done for typical farm operations. Additional field research is, therefore, needed to determine appropriate contaminant loadings for effective NMPs, and to assess whether additional treatment steps are needed. Below we discuss potential tools, treatments, and management practices that may be needed.

Computer models are valuable tools that can be used by researchers to study the transport and fate of lagoon water contaminants under various environmentally relevant scenarios that are found at NMP sites. Conventional models for contaminant transport are based on solution of mass conservation equations for flowing water (Richards equation) and solute transport (advection–dispersion equation), subject to a variety of equilibrium or kinetic reactions (e.g., Simunek et al., 2005). Improved NMPs could potentially be designed with the aid of computer models to maximize desirable effects such as nutrient uptake by plants, soil retention of pathogens, and degradation of antibiotics and hormones, and to minimize the leaching and/or runoff of contaminants.

To efficiently utilize CAFO lagoon water and diminish the potential of manure contaminants to be transported to ground water and surface water bodies, it is essential to plan and implement proper management. Manipulation of animal diet and veterinary practices are likely to be some of the most simple and cost effective ways to minimize the potential release of some contaminants into the environment. For example, it has been reported that changing cattle diet from grain to forage will reduce the numbers of pathogenic *E. coli* O157:H7 in cattle manure (Russell et al., 2000; Callaway et al., 2003). Furthermore, various approaches to deal with issues associated with antibiotic use in animal production include utilization of less antibiotics and using alternatives to antibiotics such as enzymes, competitive microbial species, precolonization of the gastrointestinal tract with favorable microbial species, and dietary additives that inhibit microbial attachment to the intestinal mucosa (Wierup, 2000; Blackman, 2000; Cook, 2004). Selection of CAFO locations away from vulnerable water resources such as shallow water tables and surface water, and on soils with relatively little structure and moderate infiltration rates, are other common sense practices to minimize the potential for water contamination. Sufficient agricultural land should also be located adjacent to CAFOs so that the manure and lagoon water that is generated may be applied to these lands at agronomic rates. Actions to further avoid water resource contamination may include: proper timing of lagoon water application (not during rainy seasons); leveling the CAFO facilities to a small incline toward a draining conduit and channeling runoff water to a designated storage reservoir (lagoon); and avoiding manure accumulation on CAFO grounds, especially during rainy seasons.

Physical separation is commonly used as an effective pretreatment method to reduce the organic load to CAFO wastewater and therefore decrease nutrient and pathogen concentrations in lagoons. Separation methods to remove organic solids from the wastewater include using stationary inclined screens, vibrating screens, rotary screens, a horizontal decanter centrifuge, hydrocyclone, as well as roller, belt, and screw presses (Zhang and Westerman, 1997). The separated manure sludge may in turn have to be treated (composted) before it can be used as a soil amendment and as a source for nutrients.

Lagoons for CAFO wastewater storage act in a similar fashion as sedimentation basins in municipal wastewater treatment. Given sufficient retention time in lagoons, significant reductions in total suspended solids are expected; that is, 35 to 65%. Most lagoons operate anaerobically. Aerated lagoons have received less attention because of their higher costs and their increased potential of transferring reactive N to the atmosphere through volatilization of NH_3. However, the potential for decreased odor and increased N reduction might increase their use if economical treatment methods emerge to mitigate NH_3 loss. Typically, CAFO wastewater storage in lagoons can reduce the amount of N by 30 to 75% through volatilization (depending on whether the lagoon is anaerobic or aerobic) (Svoboda, 1995; USEPA, 2001). Phosphorus tends to be associated with the particulate fraction of lagoon water, and N and K are usually in dissolved form. Hence, as particles settle out of solution the lagoon sludge contains less N and K but more P than the lagoon liquid. Up to 80% of the P in lagoons can accumulate in bottom sludge (MWPS, 1993; Lander et al., 1998). Heavy metals such as Cu and Zn can also accumulate in the sludge layer of lagoons (Moore et al., 2005). As for the removal of antibiotic and hormone contaminants in lagoon water, increasing the retention time of wastewater in lagoons or utilizing sequencing lagoons could be an economically feasible practice to allow sufficient time for degradation and mineralization of these chemical contaminants via abiotic and biotic mechanisms (Zheng et al., 2008). Additional

research is needed to thoroughly test this hypothesis. The bottom of CAFO lagoons should be properly sealed to prevent deep percolation of wastewater toward ground water resources. Most states have regulations regarding permissible levels of percolation rates from lagoon storage systems. For example, regulations in Kansas stipulate that percolation rates should be <0.3 to 0.63 cm d^{-1}.

Lagoon water from CAFOs may have to be adjusted to become a stable, balanced, and nutrient-rich product for reuse as irrigation water and fertilizer during implementation of NMPs. This may require active management and treatments that are not currently practiced on farms. Many municipal wastewater treatment techniques may be applied to CAFO wastewater (Zhang and Lei, 1998; Chastain et al., 2001; Vanotti et al., 2005a, 2005b; Loughrin et al., 2006). For example, chemical treatments may be used to promote flocculation of particles by neutralizing the electrostatic forces that keep them apart, causing the particles to form aggregate flakes (flocs) that rapidly settle out of solution. Commonly used flocculants include various salts of multivalent cations such as Al, Fe, Ca or Mg, and/or long-chain polymers (Dobias, 1993). Other factors such as pH, temperature, aeration, and salinity can be controlled to induce flocculation (Lindeburg, 2001). Given optimum amounts of chemical additives and sufficient settling time, flocs will form and settle out of solution to clarify wastewater. Chemical flocculation treatments involve equipment, chemicals, and maintenance that are not typically found on CAFOs. Vanotti and Hunt (1999) and Vanotti et al. (2002) estimated it would cost an additional $1.27 to $2.79 per finished hog to remove 90 to 95% of the suspended solids in swine lagoon water using chemical treatments. Additional research is needed to assess whether such approaches will be economically viable to implement.

Alternatively, it may also be possible to adapt a variety of low cost soil treatments for wastewater to remove excess amounts of organics, nutrients, pathogens, and other contaminants in CAFO lagoon water. Potential soil treatments for wastewater include: infiltration ponds and galleries, sand filtration, and subsurface flow wetlands (Schijven and Hassanizadeh, 2000; Tufenkji et al., 2002; Ray et al., 2002; Hunt et al., 2002; Weiss et al., 2005). Treatment by passage through sands can provide a physical treatment for particles in wastewater due to size limitations imposed by the pore sizes (Bradford et al., 2006a), but also provides solid–water and air–water interfaces where chemical and microbiological reactions and transformations may occur (Tufenkji et al., 2002). Furthermore, proper selection and design of the solid surface chemistry of the porous media can be used to target the retention and/or degradation of specific contaminants of interest. For example, metal oxide coatings on solid surfaces can be created to adsorb and/or inactivate a variety of microbial pathogens, as well as inorganic and organic compounds (Joshi and Chaudhuri, 1996; Ahammed and Chaudhuri, 1999). Soil treatment forms the theoretical basis for many permeable reactive barrier technologies that have been developed and applied to treat contaminated ground water plumes (Gu et al., 1999; Benner et al., 1999; Scherer et al., 2000; Waybrant et al., 2002). The collected CAFO wastewater effluent from soil treatment could in turn be used in NMPs to irrigate agricultural lands with mitigated risk to the environment.

Maintaining soil and ground water quality is the key to the long-term successful use of NMPs. Dissolved salts concentrate and accumulate in soil profiles when irrigation water that is applied to crops subsequently returns to the atmosphere via evapotranspiration. The total dissolved solids of CAFO wastewater typically range between 1500 and 3500 mg L^{-1}, equivalent to electrical conductivity levels of 2.4 to 5.5 dS m^{-1}. Salt, sodium, and osmotic effects of salinity can restrict crop establishment and growth due to changes in soil physical structure, tilth, infiltration and permeability, sodium toxicity, and salt accumulation (Ayers and Westcot, 1989; Chang et al., 1991). Selective use of gypsum, aggregate stability imparted by crops, direct seeding of crops, and careful irrigation timing of lagoon water application under a site-specific management regime are potential methods that may be used to ameliorate the negative effects of CAFO lagoon waters on soil and ground water quality (Ayers and Westcot, 1989). Salt deposits can also be flushed away by adding excess amounts of fresh water, but at a significant cost (Bouwer, 2002). Furthermore, desalination of wastewater and removing the excess salt to the ocean can cost up to $2 per 1000 L. Selection of salt-tolerant crops and timing of nutrient application are, therefore, important considerations for NMPs. The salt tolerance characteristics of various crops are provided in Ayers and Westcot (1989).

Conclusions

Concentrated animal feeding operations generate large volumes of manure-contaminated water that are typically stored in lagoons before land application. This review article discusses our current level of understanding on the environmental impact and sustainability of CAFO wastewater reuse in agriculture. Specifically, we address the source, composition, application practices, environmental issues, transport pathways, and potential treatments that are associated with CAFO wastewater.

When applied to land at agronomic rates, CAFO lagoon water has the potential to be a valuable fertilizer and soil amendment that can improve the physical condition of the soil for plant growth and reduce the demand for high quality water resources. However, excess amounts of nutrients, organics with high biochemical oxygen demand, salts, heavy metals, pathogenic microorganisms, and pharmaceutically active compounds (antibiotics and hormones) in CAFO lagoon water can all adversely impact soil and water quality, as has been well documented in the literature. The current regulatory framework is based on NMPs to ensure that lagoon water is applied to agricultural lands at agronomic rates to meet the nutrient demands of crops. Other lagoon water contaminants are implicitly assumed to be retained, inactivated, or degraded in the root zone. Additional studies are needed to thoroughly test these hypotheses.

Potential problems and weaknesses associated with the implementation of NMPs were identified in this review. One problem for proper NMP implementation is due to differences in the nutrient composition of lagoon water and the nutrient uptake rates by plants. A second more critical issue concerns the accurate estimation and delivery of water to meet plant water demands. Potential errors in water balance may occur as a result

of inaccuracies in estimates of PET, due to not accounting for all sources of plant-available water, spatial variability of soil hydraulic properties and ET, nonuniform irrigation water application, and preferential water flow that bypasses the root zone. If water in excess of PET is applied (due to irrigation inefficiency or unforeseen natural causes) to or preferential water flow occurs at specific locations on a NMP site, lagoon water contaminants may also be transported below the root zone toward ground water resources. Of special concern are contaminants that have limited ability to sorb to solid surfaces such as nitrate, or contaminants that can be associated with the mobile colloidal fraction of soil solution. Water in excess of PET will also lead to potential surface water contamination issues, especially for contaminants that are associated with sediments in runoff water such as P. If water application (from all water sources) is less than or equal to PET and preferential flow does not occur, then all lagoon water contaminants should remain in the root zone. In this case, potential environmental problems may occur due to accumulation of salts and heavy metals, incomplete degradation of antibiotics or hormones, and/or survival of pathogens. Degradation behavior for antibiotics and hormones, and survival characteristics of pathogens under field NMP conditions can be experimentally investigated, and more research is warranted on these topics.

Mitigation of the environmental risks associated with the reuse of CAFO lagoon water on agricultural lands may require planning to prevent contamination, as well as various lagoon water treatments before use in NMPs. Prevention measures may include the wise choice of locations for CAFOs, proper manure management and modification of animal diets to minimize specific contaminants (e.g., pathogens), and the use of alternatives to antibiotics. Potential treatments to CAFO lagoon water include: use of solid–liquid separators; optimization of the design of lagoon water storage basins to remove sediment loads and nutrients (aerobic compared with anaerobic); and the adaptation of various municipal wastewater treatments such as chemical flocculants and/or soil treatments.

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