

Greenhouse gas emission from wastewater irrigated soils

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Abstract

With increasing demand for world water supply, wastewater reuse is a great opportunity to meet the water need, especially for agricultural and industrial development. Wastewater originates from many sources and hence its composition differs from origin and treatment processes. Wastewater rich in organic matter acts as a soil conditioner, thereby enhancing soil health. Wastewater also acts as a source of nutrient input in agriculture which in turn can reduce, or even eliminate the need for commercial fertilisers. However, wastewater usage in agriculture poses several threats like eutrophication, salinity, toxic chemicals (heavy metal(oids), pesticides), pathogen contamination, and most notably, nutrient leaching, and greenhouse gas (GHG) emission. These threats affect public health, soil and ground water resources, environment, crop quality, ecological, and property values. Biological degradation of the organic matter present in wastewater is considered one of the anthropogenic sources of major GHGs (carbon dioxide (CO₂), nitrous oxide (N₂O), and methane (CH₄). In this paper, an overview of various sources of wastewater, effects of wastewater application on GHG emission from soil, and the strategies to mitigate wastewater-induced GHG emission from soils is presented.

Keywords: *agriculture, greenhouse gas, irrigation, mitigation, wastewater.*



1 Introduction

Wastewaters originate from a number of sources including domestic sewage, agricultural, urban and industrial effluents, and stormwater. Wastewater irrigation has many beneficial effects, including groundwater recharging and nutrient supply to plants. However, wastewater application to soil is known to increase emissions of greenhouse gas (GHG). Agricultural activities are an important source of anthropogenic GHG, contributing up to 20% of the annual emissions [1]. Carbon dioxide is the most abundant GHG in the atmosphere (360 ppmv), and is readily generated by anthropogenic activities, essentially by the burning of fossil fuels and wood. Though the atmospheric concentration of CH_4 is relatively low (1.72 ppmv), its global warming potential (GWP) is 21 times greater than that of CO_2 [1]. Methane is produced mainly from rice cultivation, anaerobic management of solid waste, biomass burning, and ruminant digestive processes. Nitrous oxide is generated by microbial activity in wastewater, soils and oceans during the degradation of nitrogen (N) rich OM, with a GWP of 310 times that of CO_2 [1, 2]. Mackenzie and Mackenzie [2] documented an increasing amount of N_2O emissions due to microbial transformation of the N contained in wastewater.

The GHG emission takes place during wastewater treatment, storage, and also when applied to the land. For example, livestock derived CH_4 represents 6–10% of total annual CH_4 emissions in the US of which significant proportion is derived from livestock wastes including manures and effluents. Wastewater is often applied to soil as it is rich in nutrients and OM. Application of organic residues to soil increases crop growth and improves soil structure. The OM in the wastewater sludge is mineralised, so the emission of CO_2 and N_2O increases from a organic waste-amended soil. The ammonium added to the soil and liberated through the mineralisation of the organic material is oxidised to nitrite (NO_2) and then to nitrate (NO_3). During this oxidation process, i.e. nitrification, N_2O is also formed. The produced NO_3 is then reduced to N_2O or N_2 under anaerobic conditions, i.e. denitrification, further contributing to GHG emissions. Application of wastewater sludge to soil can also increase emissions of CH_4 . Dissolved organic matter (DOM) in wastewaters can play a vital role on the degradation of endogenous and exogenous OM affecting C sequestration and N transformation in soils. The DOM can mediate mineralisation of organic carbon (C) into CO_2 and CH_4 as well as biological reduction of NO_3 into N_2 and N_2O . The aim of this chapter is to provide an overview on the GHG emissions from wastewater irrigated soils, and mitigation strategies to manage the GHG emissions.

2 Wastewater sources

Wastewater is derived from a number of sources that include municipal wastewater, farm wastewater, agricultural wastewater, stormwater.



2.1 Municipal wastewater

Municipal wastewater is composed of domestic and industrial wastewater. Domestic wastewater consists of discharges from households, institutions, and commercial buildings. Figure 1 shows the potential volume of domestic sewage water generated in selected countries. Land application of this wastewater depends on the country or state legislations, the crop it is applied to, and the level of treatment. Municipal wastewater contains high concentrations of nutrients, especially N and phosphorus (P), trace elements, such as iron (Fe) and manganese (Mn) and dissolved salts, particularly sodium (Na), chloride (Cl), and in some cases bicarbonates. Industrial effluents, mainly pulp and paper mill effluents are often irrigated to land after primary treatment. Pulp mill effluent has high chemical (COD) and biological oxygen demand (BOD), and some wood derived organic compounds, metal(loid)s, fatty and resin acids, and relatively high C:N ratios.

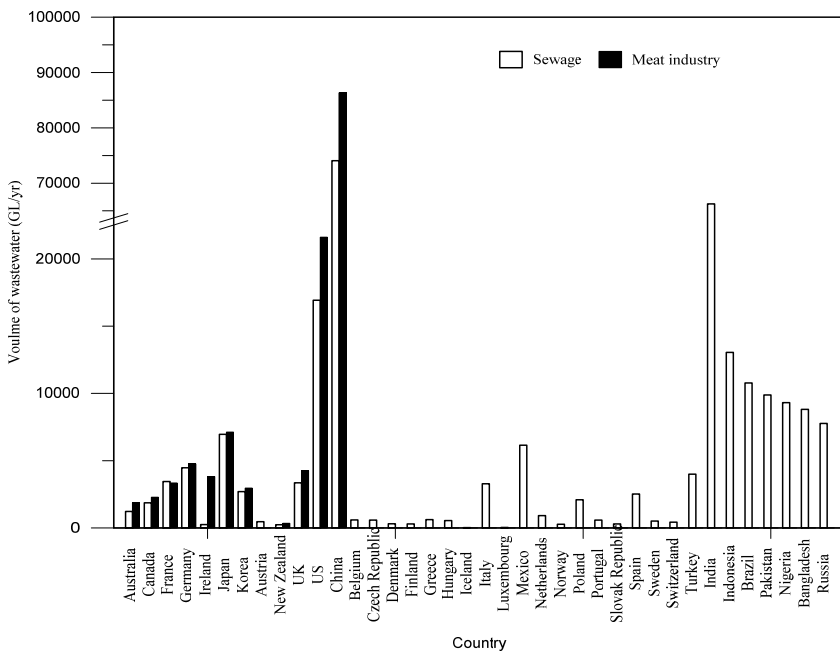


Figure 1: Potential quantities of sewage and meat industry wastewater (data not given for all countries) produced in selected countries (Water UK [3]).

2.2 Farm wastewater

Farm effluents from dairy sheds and piggeries are being increasingly employed as a source of irrigation water and nutrients in agriculture. For example, in New Zealand, dairy and piggery effluents generate annually about 9,000 Mg of N,



1,250 Mg of P and 14,000 Mg of potassium [4]. Effluents from farms differ in their composition depending on the animal production system from which they are derived (chicken, pigs, beef, and dairy). Generally, farm wastewater is rich in organic and inorganic components, and their application can increase crop yield due to the net loading of nutrients and water. In many regions, amount of farm effluents generated on a per farm basis exceeds the quantity that can be safely accommodated by the available agricultural land, and also repeated annual applications of large amounts of effluent can cause soil nutritional side effects and environmental damage.

2.3 Effluents from the agricultural industry

Recycling of agricultural industry effluents, notably effluents from animal treatment plants (fish processing plants, abattoirs) and vineyards are another common source of wastewater. For example, in Australia, agricultural drainage effluent is collected and reused as a source of irrigation water. Wastewater from intensive agricultural industries is characterised by high COD, BOD, and nutrients relative to many other wastewaters. Meat industries generate large volume of wastewater (Figure 1) and are usually disposed of to land due to high costs associated with independent treatment systems and environmental concerns over surface water discharge. Reuse of winery wastewater by grape growers and pastoralists is driven primarily through the obligations of the winery to dispose of their wastewater, preferably in a sustainable and cost-effective manner.

2.4 Stormwater

Urban stormwater harvesting has emerged in recent years as a viable option to reduce pressures on existing water sources and to alleviate adverse environmental impacts associated with stormwater run-off. This is a relatively abundant, local source of water, available throughout most urban areas. In Australia, for instance, approximately 10,300 million litres of stormwater are generated annually [5]. In many cases urban stormwater runoff contains a broad range of pollutants (pesticides, herbicides, oil, grease, and heavy metal(loids)) that are transported to natural water systems. Nutrients such as N and P are also important pollutants in stormwater. The harvesting of stormwater from industrial zones prior to its entry into natural waterways is likely to reduce the subsequent impact of point source discharge on surface waters by reducing pollutant loads.

3 Greenhouse gas emission processes

Wastewater application can lead to GHG emission by processes such as priming effect, methanogenesis, denitrification, and nitrification

3.1 Prime effect

Priming effect (PE) is the stimulation of soil organic matter decomposition by the addition of wastewater which can lead to CO₂, CH₄, and N₂O emissions. The



prime effect can be caused by the change in microbial activity. The addition of the easily available organic C triggers the activity of soil microorganisms, usually at the rate much lower than the microbial biomass [6]. When the added substrate C is low, a linear increase of extra CO₂ occurs with increasing amounts of added substrate C, resulting in a positive PE. When the amount of added substrate C is moderate, an exponential decrease in primed CO₂-C is observed. When the amount of added substrate C is more than 200% of microbial C, PE tends to be zero or negative [6]. Addition of wastewater to the soil either accelerates (positive PE) or reduces N mineralisation or immobilises added N (negative PE). Priming effect increases soil mineral N shortly after N addition and this high soil mineral N content (NH₄⁺ or NO₃⁻) can cause denitrification and hence N₂O emission.

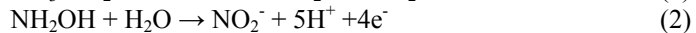
3.2 Methanogenesis

Methane production from soil is a strictly anaerobic microbial process known as methanogenesis and it requires low redox potential (Eh < - 200 mV). Methane produced in the anaerobic zones can be oxidised to CO₂ by another microbial process called methanotrophy. Methanotrophics, microorganisms involved in methanotrophy solely use CH₄ as their substrate for growth. Hence, soil serves as a source of CH₄ when the balance between CH₄ production by methanogenic bacteria and consumption by methanotrophic is positive; as a sink of CH₄ when the balance is negative [7].

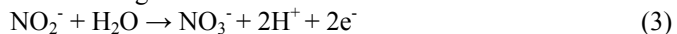
3.3 Nitrification

Nitrification is a microbial process in which reduced forms of nitrogen (NH₄) are oxidised to NO₂⁻ and subsequently to NO₃⁻. Nitrification occurs in two steps:

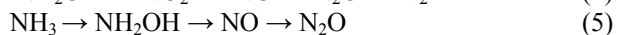
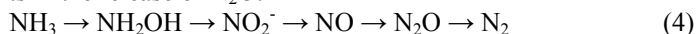
- 1) Autotrophic ammonium oxidation in which ammonia is oxidised to hydroxylamine (eqn. 1) and then to NO₂⁻ (eqn. 2) by ammonia oxidising bacteria (AOB)



- 2) Autotrophic nitrite oxidation in which NO₂⁻ is oxidised to NO₃⁻ (eqn. 3) by autotrophic nitrite oxidising bacteria



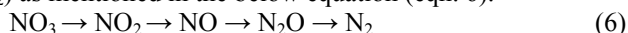
In nitrification, N₂O is produced from 2 pathways: nitrifier denitrification (eqn. 4) and hydroxylamine oxidation (eqn. 5). In nitrifier denitrification, under anaerobic or low O₂ conditions, some AOB possess nitrite reductase activity and can denitrify NO₂⁻ to nitric acid and subsequently to N₂O or N₂. Thus incomplete nitrification results in the release of N₂O.



In hydroxylamine oxidation, hydroxylamine produced from ammonia oxidation is subsequently oxidised first to nitric oxide and then reduced to N₂O.

3.4 Denitrification

Denitrification, the last step in soil N cycle is the reduction of NO_3^- to gaseous N products (NO , N_2O , N_2) as mentioned in the below equation (eqn. 6).



Denitrification is performed by a diverse group of microorganism, bacteria, archaea, and fungi, and catalysed by a range of enzymes (nitrate reductase, nitrite reductase, nitric-oxide reductase, nitrous oxide reductase). Denitrifying bacteria exhibit several reduction pathways which mainly depend on the substrate and its availability, and environmental conditions, thus resulting in the release of various products such as N_2 , or N_2O or mixture of N_2 and N_2O .

4 Greenhouse gas emissions

Wastewater contribute to GHG emission directly through the release of CO_2 , CH_4 and N_2O from C and N compounds present in the wastewater, and indirectly through their effects on soil properties thereby inducing GHG emission from soil (*e.g.* priming effect).

4.1 Carbon dioxide emission

Globally, CO_2 from fossil fuel consumption is the most significant GHG, but it represents a relatively small component of emissions from the waste sector. Carbon dioxide is emitted when organic wastes are degraded in the presence of O_2 . However, this is not considered to add to the enhanced GHG effect because organic C is part of the natural C cycle, in which photosynthesis converts CO_2 to organic matter which is subsequently converted back to CO_2 through respiration, biodigestion or combustion. The net emission of CO_2 from soil is due to two primary processes: 1) the production of CO_2 (mainly respiration by plant roots and microbes) and 2) gas transport through the soil which controls the movement of CO_2 from the soil to the atmosphere and O_2 from the atmosphere to the soil. Organic material added through treated wastewater will increase the emission of CO_2 due to decomposition. Fernández-Luqueño *et al.* [8] observed that applying urban wastewater to soil significantly increased the mean CO_2 emission rate 2.4 times ($1.74 \mu\text{gC/kg soil/h}$) compared to the unamended soil ($0.74 \mu\text{gC/kg soil/h}$), and cultivating maize further increased it 3.2 times ($5.61 \mu\text{gC/kg soil/h}$). In a similar study, Xue *et al.* [9] noticed that the cumulative CO_2 emission was significantly influenced by treated wastewater application on a silt loam soil. Large increases in CO_2 fluxes have been observed immediately following application of farm effluents to soils, being attributed to the decomposition of labile C sources [10]. Higher heterotrophic respiration rates in the presence of slurry derived C and N also contribute to the release of CO_2 from the pig slurry-applied soil [10].

4.2 Methane emission

Methane is an end product of the biological reduction of CO_2 or organic C under anaerobic conditions. Methane emissions from all surface-applied slurries and animal waste effluents are generally very high immediately after application indicating the release of entrained gas produced during the storage. Sherlock *et al.* [11] suggested that an initial burst of CH_4 soon after pig slurry application ($60 \text{ m}^3/\text{ha}$) was probably due to release of entrained gas produced in the slurry pit before application. The slurry wastewater addition appeared to restrict O_2 diffusion into the soil due to surface crust formation, thereby producing an anaerobic surface soil layer, where CH_4 could be generated. CH_4 emission from soil and volatile fatty acids in slurry are strongly correlated. Zou *et al.* [12] observed that sewage irrigated paddy cultivated land significantly increased CH_4 emission by 27% and 33% with and without chemical N addition, respectively, relative to unpolluted river water irrigation. Net CH_4 emissions from soils are determined by the difference between CH_4 formation where O_2 is absent, and CH_4 consumption where O_2 is available, and is greatly affected by the water management system (depth of flooding, draining activities). Jiang *et al.* [13] studied the effects of sheep urine and dung patches on CH_4 emission and indicated that the cumulative CH_4 emissions for dung patches, urine patches and control plots were -0.076 , -0.084 , and -0.114 g/m^2 , respectively, indicating net CH_4 sinks during the measured period. The level of CH_4 intake from urine and dung plots decreased by 25.7% and 33.3%, respectively, compared with the control plot and the cumulative CO_2 emissions increased by 0.9 and 15.9% from urine and dung plots, respectively. They observed that the sheep excrement decreased CH_4 intake and increased CO_2 emission. Tenuta *et al.* [14] suggested that grassland soils with seasonally high water tables can be significant sources of CH_4 , and the emission increased when hog slurry was applied to soils.

4.3 Nitrous oxide emission

Nitrous oxide is formed in soils during the microbiological processes of nitrification and denitrification and is highly dynamic varying with time after wastewater application and the type of application. Effluent applications may cause relatively high N_2O emissions when the soil contains NO_3 and decomposition of organic C in farm wastewater enhances N_2O emissions through both denitrification and nitrification processes [15]. In soils receiving repeated application of dairy farm effluent (DFE), frequent shifts between N_2O and N_2 production could be observed. Barton and Schipper [16] suggested that increased N_2O emissions from DFE than inorganic N fertiliser was because of enhanced denitrification activity either by increasing C availability and/or decreasing soil aeration following increased respiration. Lowering the C content of animal slurry decreased the N_2O emissions. Denitrification rates increased immediately after DFE irrigation, peaking at 24 h, and then decreased to pre-irrigation rates after 3 days [16]. Other studies indicate similar trend with N_2O emissions from three consecutive DFE irrigations, i.e. peaking within 24 hrs of application [15] but it took 1–2 weeks for the emissions to reach the level of

emissions from control treatment. These studies showed the emissions varied greatly depending on soil water filled pore space (WFPS) and the climatic conditions, and ranged from 2.04 to 5.69% of DFE-N applied. Increased soil moisture from irrigation and mineralisation of labile C and N from slurry applications favour nitrification and denitrification, resulting in highest N_2O losses after fertilisation or manure slurry application [15].

Nitrous oxide emission is found to vary with the nature of effluent irrigation, application method, and season. For example, Khan [17] measured 1.9% and 0.1%-0.3% of the applied N as N_2O emission from piggery effluent and DFE, respectively. The low N_2O emissions from DFE may be attributed to the flooded irrigation of DFE. Saturated soil water conditions can reduce N_2O emission by enhancing the complete reduction of N oxides to N_2 gas. Higher denitrification losses were found following cattle slurry injection compared to surface application to a grassland soil [15]. Liquid-waste injections have been shown to promote conditions conducive to denitrification by creating an anaerobic environment abundant in inorganic N and readily oxidizable C. Luo *et al.* [18] studied the effects of irrigating dairy-grazed grassland with FDE on N_2O emissions and observed that FDE irrigation increased N_2O emissions compared to the control and varied with changes in climatic conditions and soil WFPS. The N_2O emissions increased slightly after application of FDE, reaching a peak value soon after application in both early autumn 2004 and late summer 2005. Then the N_2O fluxes in the effluent treatment rapidly declined, and after 4–13 days were similar to those from the control.

5 Factors affecting GHG emission

GHG emission from wastewater applied soils is affected by a range of soil and environmental factors. The main factor in determining the extent of CO_2 , CH_4 and N_2O production is the amount of substrates including degradable organic matter (expressed as BOD or COD) and N in the effluent. The higher the BOD/COD content the more CH_4 and CO_2 are produced. The potential amount of CO_2 and CH_4 production following farm wastewater application to fields will depend on manure type (solid, slurry, effluent), origin (type of animal), composition, time since application, as well as climatic and soil conditions. The factors that affect CH_4 emission by soils are those that affect (1) Gas diffusion in relation with the oxydo-reduction level and CH_4 transfer, in particular the water content, the nature of clays and the type of vegetation, (2) Microbial activities in general – temperature, pH, Eh, substrate availability, physicochemical properties of soils etc, (3) Methanogenesis and in particular the competition with denitrification and sulphate-reduction and (4) Methane-mono-oxygenase activity – content in H_2 , CH_4 , NH_4^+ , NO_3^- , Cu etc.

Soil characteristics such as organic C and N content, temperature, moisture, density and porosity, are directly related to gas exchange between soil and atmosphere. Factors such as WFPS, temperature/ season, and available C and N greatly influence CO_2 flux from wastewater irrigated soils. For example, Rochette *et al.* [10] noticed that fluxes and cumulated CO_2 -C losses were greater

for spring than for fall application. They hypothesized that larger amounts of CO₂ would have solubilized in the soil solution in the fall because of higher water contents and lower temperature as compared with the spring. Urine addition to soils can result in increases in CO₂ fluxes, over and above the amounts of C applied, with the release of native soil C indicative of a priming effect [19]. Xue *et al.* [9] measured CO₂ emission from a silt loam soil irrigated with treated wastewater and noticed that the cumulative CO₂ losses showed a maximum at 60% WFPS when N fertiliser was incorporated in soils. They also observed higher CO₂ emissions in soils incubated at 60% and 80% WFPS compared to that in the drier (40% WFPS) and wetter (100% WFPS) soils. Sistani *et al.* [20] applied swine slurry to soil using various methods and observed that the method of application did not influence the CO₂ emission. This indicated that the level of tillage used to incorporate the effluent through injection or aeration did not significantly increase CO₂ emissions from no-till soils. Also they noticed that there were no significant differences in CO₂ losses, which averaged 738 and 718 gCO₂/m² in 2007 and 2008, respectively.

Nitrification and denitrification are controlled by environmental factors, cropping systems, soil management practices, inorganic or organic fertilisation, and by water regime. Temporal variations in N₂O emission can also be explained by corresponding variations in soil temperature and water content. Water-filled pore space and soil NO₃ concentrations are key factors affecting N₂O emissions. Wastewater irrigation increases WFPS and a WFPS above 60% increases N₂O losses due to denitrification [21]. Effluent applications enhance microbial activity, which reduces soil O₂ levels, creating conditions that favour N₂O emissions [15]. Carbon and N present in wastewater strongly influence N₂O emission. Amon *et al.* [22] reported a 25% reduction in N₂O emissions following the addition of anaerobically digested manure slurries compared with undigested liquid manure due to lower concentrations of labile C and N in the digested effluents. Zou *et al.* [12] noticed that relative to river water irrigation, sewage irrigation increased N₂O by 68% and 170% from paddy fields with and without N application, respectively. In contrast to the above mentioned studies which highlight the importance of denitrification process in N₂O emission, Master *et al.* [23] reported that reclaimed effluent application did not increase the N₂O emission. They suggested that the effluent treatment level (BOD₅=100 mg/L) may have been low enough not to affect the N₂O emissions in the short term

6 Mitigation strategies

Several management practices and technologies help mitigate GHG emissions from wastewater treated soils and, wastewater treatment and storage units. Reduced tillage could increase soil organic matter, enhance soil water-holding capacity and reduce the need for irrigation water. Increased soil organic matter could also improve natural soil fertility, thereby decreasing the need for inorganic fertilisers and organic amendments. Options to reduce the amount of excreta N produced, whilst maintaining total productivity, such as low N diets



for dairy and intensive beef cattle, could yield a 15% reduction in N_2O emissions. Options that increase the N use efficiency of excreta or fertiliser, such as the strategic use of stand-off pads or nitrification inhibitors, could reduce N_2O emissions by up to 20%. Improving the drainage of poorly or imperfectly draining soils, or avoiding soil compaction on these soils, is estimated to reduce N_2O emissions by 7–10%. In terms of the advanced fertilisation techniques, the use of nitrification inhibitors and/or slow release fertilisers has been shown to reduce N_2O emissions substantially. Non-leguminous cover crops are efficient scavengers of residual soil NO_3 , thereby reducing leaching losses. Jarecki *et al.* [24] investigated the cover crop effects on N_2O emission from a liquid swine manure-treated Mollisol. They observed a significant reduction in N_2O emissions in the presence of rye. Luo *et al.* [18] suggested that strategic application of FDE during dry summer and autumn seasons could reduce N_2O emissions and delaying effluent irrigation after grazing events could further reduce N_2O emissions by reducing the levels of surplus mineral-N. Drainage system influence CH_4 emission as well as N_2O emission from waste water irrigated rice field. Multiple drainage and mid-season drainage at flowering period can help mitigate CH_4 emission. Mid-season drainage reduced CH_4 emissions by 43% because the flux of O_2 into the soil created aerobic conditions, unfavourable to methanogenic bacterial activity [25]. It should be noted that in the aerobic zones of wetland and upland soils, CH_4 is oxidised into CO_2 by methanotrophs [15].

7 Summary and conclusions

Growing population, increased urbanisation, improved living conditions and economic development have led to a considerable increase in the volume of wastewater generated by domestic, industrial and commercial practices. The use of treated wastewater for both agricultural production and environmental protection has increased in recent years in several continents including Australia, Europe and North America. Adequate management of wastewater irrigated soils and crops results in a reduction of GHG emissions by storing atmospheric C as soil organic matter. Though major sources and factors controlling trace gas emissions from wastewaters and waste management systems are well known, further research and development is needed to improve the accuracy and utility of these tools.

Acknowledgement

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