Chapter 16

Decentralised wastewater treatment and reuse plants: Understanding their fugitive greenhouse gas emissions and environmental footprint

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16.1 INTRODUCTION

Centralised urban water and wastewater systems serving large urban areas are rapidly being subjected to a growing number of operational challenges, primarily due to increasing urbanisation and ongoing population growth (Sharma *et al.* 2010a). To accommodate this population growth, urban densities are increasing and urban boundaries are expanding. Centralised systems have provided considerable benefits to the general public, via the provision of reliable water treatment services alongside increased health and sanitation. However, due to the increased demand on water resources, the aging and refurbishment needs of our infrastructure, as well as the need to minimise contaminant loads to receiving environments, questions have been raised regarding the long term viability of conventional centralised solutions for providing ongoing water treatment services. Moreover, some new urban areas are developing in close proximity to environmentally sensitive areas, where wastewater cannot be discharged through conventional approaches. Also, not all of the new developments can be connected to conventional centralised systems due to long distance transportation needs for bringing freshwater in and taking wastewater out, which makes these approaches economically difficult (Sharma *et al.* 2010b).

In order to meet these challenges, decentralised water and wastewater reuse systems are being implemented either in combination with centralised systems or as stand-alone systems (Sharma *et al.* 2010b; Sharma *et al.* 2013a). Decentralised reuse systems are planned using Integrated Urban Water Management (IUWM) and Water Sensitive Urban Design (WSUD) approaches, which provide the potential to implement local water systems by considering local requirements (Sharma *et al.* 2013b). These approaches can also offer the opportunity to use local water sources and close the loop on waste streams through taking a ‘fit for purpose’ approach, matching the quality of source water to the quality requirements of their end-use. Separate collection and treatment of various waste streams and recovery of valuable water, nutrients and energy is also possible through these systems (Wilderer, 2001).

As decentralised reuse systems are comparatively novel compared to conventional centralised approaches, the understanding and knowledge of these systems is still being developed in regards to planning, design, implementation, operation and maintenance, health impacts and environmental impacts (Sharma *et al.* 2012). Therefore knowledge gaps exist in selecting suitable servicing options. For decentralised wastewater treatment and reuse systems, limited information is available on the total environmental footprint related to their day to day operation, in particular the amount of non carbon dioxide (CO_2) fugitive greenhouse gas (GHG) emissions that they release (Sharma *et al.* 2009). This is very important to quantify, as it is necessary to determine if the increasing installation of decentralised reuse systems will be environmentally sustainable over an extended time period. This chapter details a short-term pilot study that attempts to begin to address these knowledge gaps related to the emission of fugitive GHGs, by analysing the emissions from a cross-section of decentralised wastewater reuse systems. It is believed that the outcomes from this research will assist water professionals in better understanding the potential environmental footprint of decentralised wastewater reuse systems and consequently may also help to guide water professionals in selecting appropriate decentralised wastewater reuse systems in the future.

16.2 EMISSION MECHANICS OF N_2O AND CH_4 FROM WASTEWATER TREATMENT SYSTEMS

Fugitive emissions of the GHGs, mainly methane (CH_4) and nitrous oxide (N_2O) are known to be produced and released by various anaerobic and aerobic wastewater treatment processes. Once released into the atmosphere, N_2O and CH_4 are much more effective at trapping heat in comparison to CO_2. More precisely, N_2O has 298 times greater atmospheric heating potential over 100 years in comparison to CO_2 and CH_4 has 25 times greater atmospheric heating potential over 100 years in comparison to CO_2 (Intergovernmental Panel on Climate Change, 2007). Additionally, N_2O can breakdown and eliminate ozone (O_3) in the stratosphere (Ravishankara *et al.* 2009). This has a negative impact upon the Earth's ecosystem, as the reduction of O_3 results in greater levels of biologically damaging downwelling solar UV radiation (UV_{BE}) being able to penetrate the atmosphere and reach the surface of the Earth.

In the activated sludge wastewater treatment process, N_2O is generated as a by-product of the autotrophic nitrification and heterotrophic denitrification processes (Law *et al.* 2012). Specifically, nitrification takes place under aerobic conditions when ammonium-oxidizing bacteria (AOB) and ammonium-oxidizing archaea (AOA) convert ammonia into nitrite, and nitrite-oxidizing bacteria (NOB) converts nitrite into nitrate (Kampschreur *et al.* 2009). In wastewater treatment processes, nitrification is most likely to be initiated by autotrophic AOB and NOB using either ammonia or nitrite as a source of energy and CO_2 as a source of carbon (Kampschreur *et al.* 2009). AOB have been shown to produce N_2O, as they contain the enzymes to breakdown NO_2^--N and NO leaving N_2O as a remainder (Global Water Research Commission, 2011). Denitrification occurs in anaerobic conditions and is facilitated metabolically by a large range of bacteria, archaea and micro-organisms that couple the oxidation of organic or inorganic substrates to the reduction of nitrate, nitrite, NO and N_2O (Global Water Research Commission, 2011).

Since N_2O is generated as an intermediate product during denitrification, incomplete denitrification may result in the release of N_2O into the surrounding environment. The factors most closely associated to the generation of N_2O during nitrification and denitrification are still not completely understood. However, N_2O production may be correlated to a number of wastewater treatment plant (WWTP) operating parameters such as dissolved oxygen concentration and various mass transfer/solids retention conditions. Recent studies performed at centralised WWTPs indicate that levels of dissolved oxygen, ammonia and nitrite can be used as predictors of the extent of N_2O emission taking place in treatment reactors and that nitrification is generally a higher contributor to N_2O output in comparison to denitrification (Rassamee *et al.* 2011; Ho Ahn *et al.* 2010). In addition, N_2O emissions measured from aerated treatment processes are generally much higher than those measured from non-aerated treatment processes (Rassamee *et al.* 2011; Ho Ahn *et al.* 2010). Sampling at centralised WWTPs is showing that the N to N_2O conversion percentage can vary extensively from WWTP to WWTP depending on a wide variety of process parameters (Kampschreur *et al.* 2009; Foley *et al.* 2010; Townsend-Small *et al.* 2011; Global Water Research Commission, 2011) and could be greatly dependent upon the nutrient loading existent in the influent, which can vary extensively throughout a short time period. Recent diurnal N_2O measurements are showing that a peak in N_2O generation and emissions occurs during the interval over which the maximum daily N loading arrives into a WWTP (Lotito *et al.* 2012).

In a typical wastewater treatment system, CH_4 is produced predominantly via the anaerobic decomposition of organic matter by methanogenic bacteria. This process is generally referred to as anaerobic digestion. In some cases, aerobic wastewater treatment processes can require more oxygen than is delivered via diffusion, and as a result, when surplus mechanical aeration is not available, methanogenic bacteria begin the anaerobic digestion process from which CH_4 is generated and released (Czepiel *et al.* 1993). The anaerobic digestion process occurs over four stages, with each stage requiring a specific group of micro-organisms to initialise: (i) *Hydrolysis*: the breakdown of non-soluble biopolymers to soluble organic compounds; (ii) *Acidogenesis*: the breakdown of soluble organic compounds to CO_2 and volatile fatty acids; (iii) *Acetogenesis*: the breakdown of volatile fatty acids to H_2 and acetate; and (iv) *Methanogenesis*: the breakdown of acetate, CO_2 and H_2 to CH_4 (Mes *et al.* 2004). The rate and extent of anaerobic digestion and CH_4 production are both positively correlated to temperature, toxicity, pH and chemical oxygen demand (COD). COD is used as a direct indictor to predict the potential of biogas emission to occur from a wastewater sample (Mes *et al.* 2004). CH_4 can still be emitted from aeration tanks, even when a high oxygen concentration is present. This is possible as most CH_4 is produced earlier in the sewer pipeline system adjoining the WWTP or it is delivered via rejection water released from sludge handling processes (Global Water Research Commission, 2011). The CH_4 that arrives into aeration tanks is released into the atmosphere via gas stripping.

16.2.1 Study specification and objectives

Both CH_4 and N_2O emissions from centralised WWTPs have been well documented and quantified by the application of a variety of online gas analysis instrumentation and grab sampling techniques. Some examples of these studies include Czepiel *et al.* (1993), Czepiel *et al.* (1995), Ho Ahn *et al.* (2010), Global Water Research Commission (2011), Townsend-Small *et al.* (2011) and Winter *et al.* (2012). Conversely, there is limited information available on the temporal and spatial distribution of the fugitive emissions released at decentralised reuse systems treating smaller daily volumes of sewage. As such, real-world CH_4 and N_2O emission studies carried out at small-scale decentralised reuse systems are necessary in order to resolve this gap in the knowledge base. In order to better ascertain the amount of fugitive CH_4 and N_2O emissions produced by decentralised reuse systems, a series of CH_4 and N_2O gas flux measurements were performed at three different Australian decentralised reuse systems over the months of spring in 2012 using an online non-dispersive infrared (NDIR) gas analyser combined with a gas capture flux hood. This chapter details the extent of the CH_4 and N_2O emissions measured throughout this research campaign and directly compares these results to emissions estimates calculated from a wastewater GHG emission model that is currently employed by Australian wastewater treatment operators to evaluate the annual environmental footprint of their systems. The model is a series of analytical equations that has been developed by experts working in consultation with the Australian Government, but is not commercially available. A complete description of the CH_4 and N_2O components of the model is included in Section 16.3.3.

16.3 MEASUREMENT CAMPAIGN SPECIFICATION AND ANALYSIS METHODOLOGIES

16.3.1 Reuse systems specifications

For the purposes of this research, a decentralised reuse system is defined as a wastewater treatment system managing influent from a population with no greater than 75,000 people with a daily flow rate of no more than 5×10^6 m^3/year. To acquire an inventory of fugitive N_2O and CH_4 emissions data, a series of field investigations were carried out over the months of spring at three decentralised reuse systems situated over the greater metropolitan area of a large Australian city (Melbourne, Victoria, 37°48′49″S 144°47′57″ E, Altitude: 31 m). Off-gases from one reuse system with a large catchment population (Site A), one reuse system with a small catchment population (Site B) and one sewer mining facility used for irrigation over an 18-hole golf course (Site C) were analysed. All three of these reuse systems use the activated sludge treatment process. The reuse systems were chosen due to their varying age, spatial footprint, catchment area, treatment regime and daily average inflow and organic loading, in order to provide a generally representative cross-section of the types of decentralised reuse systems currently working in countries with modern sewage treatment infrastructure. Table 16.1 provides the operational data and calculated influent water quality parameters for each of the three reuse systems evaluated in this study. Generalised treatment regime schematics for the three reuse systems have been presented previously in Schouten *et al.* (2013a) and Schouten *et al.* (2013b).

Table 16.1 Decentralised reuse system operational metadata and water quality parameters.

	Site A	**Site B**	**Site C**
Function	Wastewater treatment for local community	Wastewater treatment for local community	Sewer mining for golf course irrigation
Effluent reuse application	Irrigation of local vineyards, tree plantations and on-site lawns	Redistribution into river system for reuse by local farmers and businesses	Irrigation of greens, fairways, lawns and gardens around the golf course
Predominant influent type	Domestic	Domestic	Domestic
Catchment Population (approximate)	75,000	400	3000
Yearly COD mass load (metric tonnes)	2660	25	88
Yearly BOD mass load (metric tonnes)	1662	9.4	46.4
Yearly total N mass load (metric tonnes)	335	1.7	11
Yearly inflow (m^3) (Daily flow rate (m^3/ day))	4.86×10^6 (13.3×10^3)	21×10^3 (57.5)	1×10^5 (274)

Each site extensively reuses its treated effluent. Site A reuses a sizeable volume of its treated effluent to irrigate local vineyards and tree plantations. The treated effluent is also used to water lawns around the treatment systems and the on-site control office. In addition, Site A collects a stockpile of dried digested sludge for use as fertiliser on nearby lawns and gardens. Site B sends its treated effluent back into a local river system, from which it can be readily collected and used by local farmers, horticulturalists and businesses for irrigation purposes. Site C recycles all of its treated effluent to continuously irrigate the fairways and greens on each hole at the golf course. The treated effluent produced at Site C is also used to water the gardens and lawns around the greens and the clubhouse.

Fugitive N_2O and CH_4 fluxes were measured only from the aeration tank systems at Site A, Site B and Site C, as at these sites aeration tanks had the most sizeable spatial footprint and were completely atmospherically exposed, and consequently had the most potential to release the highest cumulative emissions. N_2O and CH_4 fluxes were measured simultaneously at evenly spaced positions across the length of each aeration tank in order to evaluate and quantify the spatial distribution of the gas emissions. The measurements were performed over a single day during spring (September 2012 to November 2012 in the Southern Hemisphere) in order to obtain a general ‘snapshot’ of the fugitive emissions produced at each site. Table 16.2 displays the average values and standard deviations (1σ) for water quality parameters measured at the reuse systems during the measurement campaign along with the averaged N_2O and CH_4 fluxes recorded during the sampling interval.

Table 16.2 Reuse system peripheral water quality data and averaged CH_4 and N_2O fugitive emissions for Site A, Site B and Site C.

	Dissolved oxygen (mg/l) (Mean, ±Std. Dev)	**pH (Mean, ±Std. Dev)**	**Conductiv. (μS/cm) (Mean, ±Std. Dev)**	**Sewage temperature (°C) (Mean, ±Std. Dev)**	**CH_4 (g CH_4/m^2/d) (Mean, ±Std. Dev)**	**N_2O (g N_2O/m^2/d) (Mean, ±Std. Dev)**	**Number of Measurements (N)**
Site A Aeration tank *Spring*	0.94 (±0.28)	7.14 (±0.17)	994.8 (±36.03)	18.4 (±0.05)	2.4 (±3.4)	1.09 (±0.8)	18
Site B Aeration tank *Spring*	0.099 (±0.05)	6.7 (±0.02)	470.6 (±6.06)	18.3 (±0)	0.07 (±0.02)	0 (NA)	18
Site C Aeration tank *Spring*	1.29 (±0.39)	6.4 (±0.08)	705.2 (±11.1)	22.5 (±0.14)	0.22 (±0.16)	0.71 (±0.25)	18

16.3.2 Gas analysis instrumentation and sampling technique

The gas sampling carried out through the measurement campaign followed a methodology similar to the gas sampling procedures used by Tremblay *et al.* (2004) and Carignan (1998). In-situ gas capture was made on the sewage surface with a commercially available buoyant airtight flux hood (St Croix Sensory Inc., United States) connected to a primary standard calibrated (traceable to the National Institute of Standards and Technology) NDIR gas analyser (Horiba Ltd., Japan). Emitted gases were trapped in the flux hood and sent to a gas conditioning unit (Horiba Ltd., Japan) via a pump operating at a constant flow rate. The gas conditioning system removed water vapour, acids and other pollutants from the gas flow before it entered the NDIR gas analyser. After the CH_4 and N_2O gas concentrations had been calculated by the NDIR gas analyser, the sampled gases were returned into the flux hood so they could mix continuously, enabling a more accurate estimate of gas concentration to be obtained over time (Lambert & Frechette, 2005). Generally, for gas measurements in remote or isolated field locations (such as decentralised WWTPs), in-situ gas collection with a gas capture hood combined with a NDIR gas analyser is the most appropriate. This is due to its relative ease of use, portability and rapid flux calculation capability.

Gas concentrations were recorded over 10 (±5) minute intervals to reduce the influence of the chamber effect (Venterea *et al.* 2009). The flux hood was held in place with a tightened rope to minimise the effect of wave action and turbulence occurring on the sewage surface and to keep the flux hood in the same position over the measurement interval. Following each measurement, the flux hood was removed from the sewage surface so that gas concentrations could return to ambient levels before the next measurement. The gas concentration data from each reuse system was logged continuously to a laptop computer beginning at 10:00 AM (±2 hours) until 2:30 PM (±2 hours) during each day of sampling. From this sampling regime, trends in gas flux could be readily quantified over the time of day at which the influent flow rate and nutrient loading was most likely to be at its highest value. Figure 16.1a depicts the deployment of the flux hood on top of the Site C aeration tank and Figure 16.1b displays the operation of the NDIR gas analyser, the gas conditioner and the laptop computer over the Site B aeration tank. The method described by Tremblay *et al.* (2004) was used to calculate CH_4 and N_2O gas flux. For this, a linear regression was applied separately to the CH_4 and N_2O gas concentration data recorded over the sampling time during each particular measurement interval. From the gas concentration data, CH_4 and N_2O gas flux could be calculated directly using Equation 16.1 (Tremblay *et al.* 2004):

$$\text{Flux} = \frac{m \times V \times \alpha \times \beta}{A \times \gamma} \tag{16.1}$$

where m is the slope from the linear regression set to the gas concentration data over the sampling time (ppm/second); V is the volume under the flux hood (m^3); α is a gas concentration conversion factor (for CH_4: 655.47 μg/m^3/ppm; for N_2O: 1798.56 μg/m^3/ppm); β is a temporal conversion factor (86,400 seconds/day); A is the area under the flux hood (m^2) and γ is a magnitude conversion factor (1×10^6 μg/g). Flux is given in g/m^2/day.

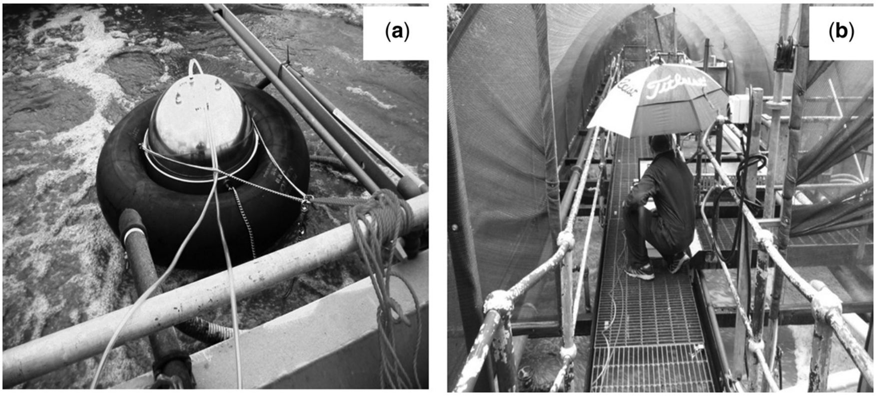

Figure 16.1 Example deployment of the gas analysis system in the field: (a) The flux hood on top of the Site C aeration tank; (b) The gas analysis and data collection workstation being worked on over the Site B aeration tank.

Continuous measurement of various water quality parameters were made simultaneously to the gas flux measurements using a water quality sonde (Aquaread Ltd., United Kingdom), with the sonde positioned in close proximity to the flux hood. The sonde was lowered to a depth of approximately 0.5 to 1 m under the sewage surface during sampling. The water quality parameters that were measured were temperature, pH, dissolved oxygen and conductivity. At the start of each site visit, the water quality sonde was calibrated to a standard calibration solution formulated by the sonde manufacturers (Aquaread Ltd., United Kingdom). More water quality and daily operational data such as influent flow rate, effluent flow rate, influent total N mass load, influent total COD and BOD mass load, oxygen delivery profiles and inflow pump timings were obtained from the site engineers when necessary.

To calculate a basic estimate of the total percentage conversion of influent nutrient loading to CH_4 and N_2O gas taking place at each decentralised system, basic emission factor calculations were performed. To do this, the mean fugitive N_2O emission measured over spring was normalised to the total annual $N_{INFLUENT}$, and the mean CH_4 emission recorded over spring was normalised to the total annual $COD_{INFLUENT}$ for the aeration tanks at Site A, Site B and Site C. These normalisations were made using Equation 16.2 and 16.3:

$$N_2OEF = \left[\frac{(N_2O)_{MEAN}}{(N_{INFLUENT})_{ANNUALTOTAL}}\right] \times 100\% \tag{16.2}$$

$$CH_4EF = \left[\frac{(CH_4)_{MEAN}}{(COD_{INFLUENT})_{ANNUALTOTAL}}\right] \times 100\% \tag{16.3}$$

where: N_2OEF and CH_4EF are the annual emission factors for N_2O and CH_4 respectively; $(N_2O)_{MEAN}$ is the mean of the N_2O flux measurements (tonnes) integrated over the tank surface area made over spring; $(N_{INFLUENT})_{ANNUALTOTAL}$ is the annual total N (tonnes) arriving in the influent at each system; $(CH_4)_{MEAN}$ is the mean of the CH_4 flux measurements (tonnes) integrated over the tank surface area made over spring and $(COD_{INFLUENT})_{ANNUALTOTAL}$ (tonnes) is the annual total COD arriving in the influent at each system.

16.3.3 Wastewater GHG emissions modelling

The cost and time required to set up and maintain online gas measurement instrumentation at both centralised and decentralised WWTPs is often highly prohibitive. Consequently, semi-empirical modelling techniques are predominantly used by wastewater treatment operators to estimate the amount of fugitive N_2O and CH_4 emissions being released from their wastewater treatment systems each year. These models generally employ input data obtained from real-world continuous or intermittent measurements made using online or handheld water quality instrumentation, from wastewater grab samples analysed using chemical or optical methods in a laboratory, or from inferences or extrapolations taken from previously published investigations. Currently, in Australia a large number of wastewater treatment operators are required to report on the total N_2O and CH_4 emissions produced annually by their WWTPs as part of the National Greenhouse Energy Reporting Scheme (NGERS). NGERS currently uses two separate models to estimate direct N_2O and CH_4 emissions released during wastewater treatment. A simplified version of the NGERS N_2O emission model applicable to this research is as follows (Global Water Research Commission, 2011):

$$N_2O_{WWT} = \left[(N_{IN} - N_{OUT}) \times EF_{N_2O_{WWT}}\right] + \left[N_{OUT} \times EF_{N_2OD}\right] \tag{16.4}$$

where N_2O_{WWT} is the amount of N_2O gas emitted from wastewater (tonnes $CO_{2EQUIVALENT}$), N_{IN} is the amount of nitrogen entering the WWTP/system (tonnes), N_{OUT} is the amount of nitrogen leaving the WWTP/system to re-enter the local environment (tonnes), $EF_{N_2O_{WWT}}$ is the default N_2O emission factor for domestic wastewater treatment (4.9 tonnes $CO_{2EQUIVALENT}$/tonnes $N_{REMOVED}$) and EF_{N_2OD} is the default N_2O emission factor for treated wastewater discharge (4.9 tonnes $CO_{2EQUIVALENT}$/tonnes $N_{REMOVED}$). $EF_{N_2O_{WWT}}$ is always set to zero for WWTPs that do not have secondary nitrification-denitrification treatment in place.

The basic NGERS CH_4 emission model used to predict fugitive CH_4 emissions from wastewater treatment is given below (Global Water Research Commission, 2011):

$$CH_{4WWT} = \left[\left(COD_{IN} - COD_{SLUDGE} - COD_{OUT}\right) \times EF_{CH4WWT} \times F_{ANAEROBIC}\right] \quad (16.5)$$

where CH_{4WWT} is the quantity of CH_4 gas emitted from wastewater (tonnes $CO_{2EQUIVALENT}$), COD_{IN} is the amount of COD coming into the WWTP/system (tonnes), COD_{SLUDGE} is the amount of COD removed in sludge and treated in the WWTP/system (tonnes), COD_{OUT} is the quantity of COD exiting the WWTP/system, EF_{CH4WWT} is the default CH_4 emission factor for domestic wastewater treatment (5.3 tonnes $CO_{2EQUIVALENT}$/tonnes $COD_{REMOVED}$) and $F_{ANAEROBIC}$ is the fraction of COD treated anaerobically within the WWTP/system each year. For all WWTPs running a managed aerobic treatment processes, $F_{ANAEROBIC}$ is automatically set to zero.

Emissions estimations made by the NGERS models (Equations 16.4 and 16.5) were directly evaluated against the measured N_2O and CH_4 emissions data collected at Site A, Site B and Site C over the March 2012 to April 2013 measurement campaign period. These comparisons are discussed in further detail in Section 16.4.1.

16.4 MEASUREMENT CAMPAIGN RESULTS AND DISCUSSION

16.4.1 Fugitive emissions

Figures 16.2 (a, b and c) display the N_2O and CH_4 emissions measurements taken from the aeration tanks over a single day of sampling at the three sites. In the Site A data displayed in Figure 16.2a, six emissions measurements were obtained in one section of the aeration tank from 9:49 AM to 10:45 AM. Following this, the flux hood was moved approximately five metres lengthways across the tank where a further six emissions measurements were performed from 11:02 AM to 11:55 AM. After this, the flux hood was moved ten metres lengthways once again where a final series of six emissions measurements were taken from 12:09 PM to 1:07 PM. In the Site C data shown in Figure 16.2c emissions measurements were made in six separate sections across the aeration tank, each spaced approximately one metre apart. Each of these sections was evaluated over the following intervals: 10:41 AM to 10:57 AM (section 1), 11:04 AM to 11:25 AM (Section 2), 11:37 AM to 11:56 AM (section 3), 12:05 PM to 12:22 PM (Section 4), 12:35 PM to 12:52 PM (Section 5) and 1:03 PM to 1:24 PM (Section 6). Due to limited available space at Site B, flux measurements were recorded in same position on the aeration tank surface over the single day sampling period.

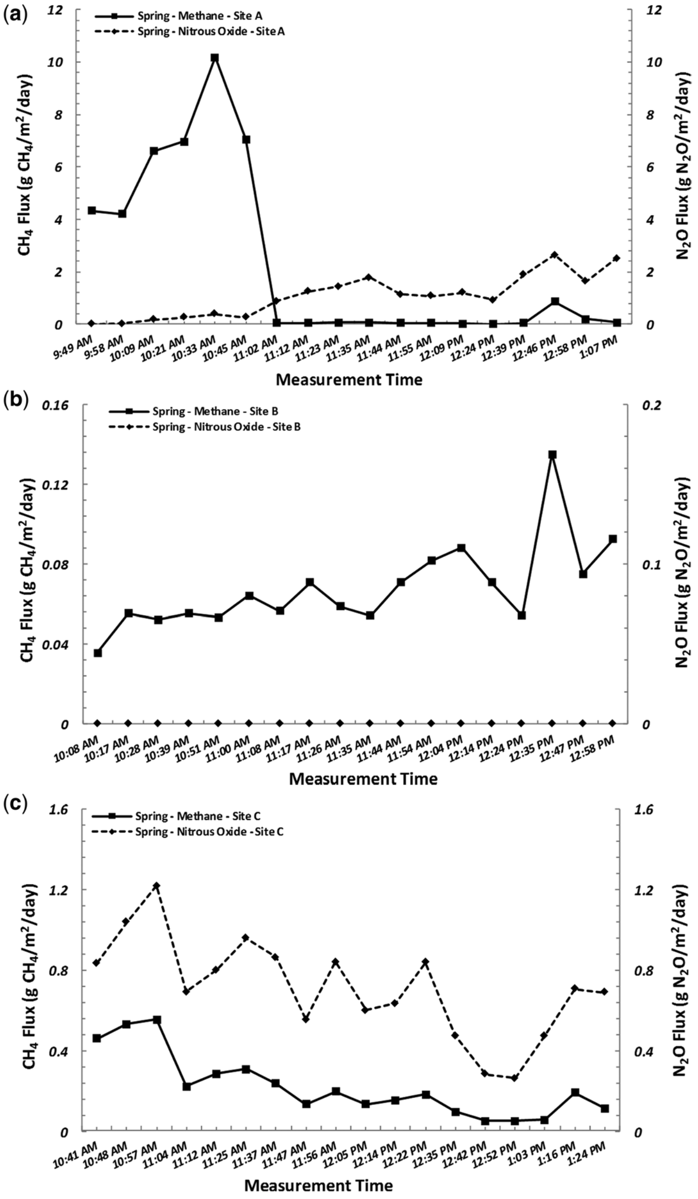

Figure 16.2 (a) N_2O and CH_4 emissions measurements taken from the aeration tanks made over a single day of sampling at Site A. (b) N_2O and CH_4 emissions measurements taken from the aeration tanks made over a single day of sampling at Site B. (c) N_2O and CH_4 emissions measurements taken from the aeration tanks made over a single day of sampling at Site C.

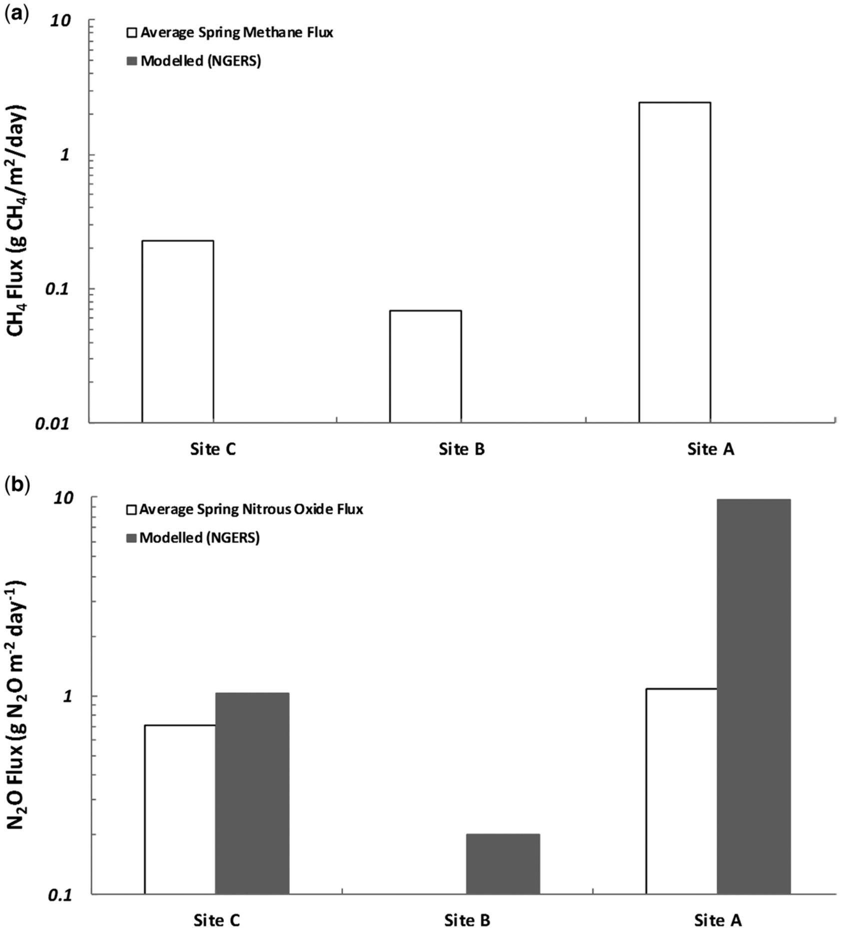

Figure 16.3 (a) Daily averaged CH_4 emissions data measured from the aeration tanks located at Site A, Site B and Site C over spring 2012. The measured CH_4 data is compared to CH_4 emissions estimates calculated by the NGERS models. (b) Daily averaged N_2O emissions data measured at the Site A, Site B and Site C aeration tanks over spring 2012. The measured N_2O data is compared to N_2O emissions estimates calculated by the NGERS models.

From these staggered emissions measurements, significant variations in the spatial (lengthways across the tank) and short-term temporal (with measurements being made in the same position) distributions of CH_4 and N_2O emissions were found to occur at Site A, Site B and Site C. Some of the most extensive temporal variations in CH_4 flux found were: (i) a 32% increase (7 g CH_4/m^2/day to 10.2 g CH_4/m^2/day) over a time interval of twelve minutes at Site A in section 1; (ii) an increase of gas output by a factor of close to 2½ (0.054 g CH_4/m^2/day to 0.13 g CH_4/m^2/day) over ten minutes at Site B; and (iii) a tripling of flux (0.061 g CH_4/m^2/day to 0.19 g CH_4/m^2/day) over thirteen minutes at Site C in section 6. N_2O flux was also found to fluctuate by substantial amounts over short time periods, with the most prevalent instances being a doubling of flux (0.94 g N_2O/m^2/day to 1.89 g N_2O/m^2/day) taking place over a time interval of 15 minutes at Site A in section 3, and a decrease of 36% (0.87 g N_2O/m^2/day to 0.56 g N_2O/m^2/day) over ten minutes at Site C in section 3. Additionally, N_2O fluxes measured in quick succession (<10 minutes apart) over a distance of no greater than five metres were found to increase by as much as a factor of three at Site A (0.27 g N_2O/m^2/day to 0.89 g N_2O/m^2/day between section 1 and section 2) and could decrease by as much as 43% at Site C (1.2 g N_2O/m^2/day to 0.7 g N_2O/m^2/day between section 1 and section 2). Similar levels of high variability were also measured for CH_4 fluxes measured over the same distances at Site A and Site C.

As mentioned in the previous paragraph, spatial distributions of CH_4 and N_2O flux were not obtained at Site B due to the small aeration tank surface area over which there was minimal space to reposition the flux hood. However, it could be assumed that similar substantial spatial variations in both CH_4 and N_2O flux may also take place there. It is difficult to pinpoint the exact cause of these considerable variations in the spatial and short-term temporal distribution of CH_4 and N_2O fluxes at each of the decentralised reuse systems. The spatial variations may be linked to oscillations in the amount of organic content and dissolved gas entering the aeration tank during the measurement interval. As a result of variability in the inflow organic and dissolved gas loading, the amount of biological activity and gas available for stripping within a given volume at one section of the tank may be completely different to another section of the tank. Fluctuations in flux output measured over time and across the surface area of an aeration tank could also be due to gradual changes occurring to the chemistry of the mixed liquor located within different positions. These changes are most likely to be facilitated by changes in total aeration delivery and the positioning of the bubblers/oxygen supply in the tank aeration system.

Figure 16.3a and Figure 16.3b show respectively the daily averaged CH_4 and N_2O emissions data recorded from the aeration tanks located at Site A, Site B and Site C over spring. In these two figures, the measured CH_4 and N_2O emissions data is compared directly to CH_4 and N_2O emissions estimates calculated by the NGERS modelling framework specified in Section 16.3.3. From Figure 16.3a it can be seen that Site A emitted the highest amount of CH_4 on average in comparison to Site B and Site C, with Site A producing peak CH_4 emissions of 2.4 g CH_4/m^2/day. This was expected as Site A treats a far greater daily loading of COD in comparison to Site C and Site B and as such has the highest potential for anaerobic digestion to occur throughout its treatment regime. Site C released the next highest amount of CH_4, that being 0.22 g CH_4/m^2/day and Site B emitted the lowest CH_4 level of 0.07 g CH_4/m^2/day. In Figure 16.3b, it is clear that Site A also emitted a higher average amount of N_2O gas in comparison to Site B and Site C. Specifically, Site A emitted an average N_2O emission output of 1.09 g $N_2O/m^2/day$, in comparison to 0.71 g $N_2O/m^2/day$ from Site C and 0 g $N_2O/m^2/day$ from Site B. Once again, this outcome was anticipated to occur as Site A takes in a much larger volume of N in its influent flow every day compared to Site C and Site B. Subsequently, of all the three sites, Site A has by far the greatest potential for larger amounts of N_2O gas conversion to take place during nitrification and denitrification processes occurring within its treatment system. Despite its much lower yearly inflow and total N mass load, Site C still managed to emit close to 65% of the total average N_2O gas output delivered by Site A. It is unclear why Site C was emitting this substantial amount of N_2O. However, this disproportionate output may be occurring due to periods of inadequate aeration (as a result of an inefficient aeration system) or could be due to large peaks in ammonium entering the system, which can lead to an increase in N_2O generation and its subsequent release (Global Water Research Commission, 2011).

It is clear from Figure 16.3a that the NGERS CH_4 emissions model (Equation 16.5) predicts that no CH_4 emissions should occur from the aeration tanks at Site A, Site B and Site C. This is due to the initial assumption that a managed aerobic treatment process does not treat any organic loading anaerobically and as such does not have the potential to emit any fugitive CH_4 gas. This assumption effectively sets the $F_{ANAEROBIC}$ term in the NGERS CH_4 emissions model to zero. However, dissolved CH_4 gas can still enter aerobic treatment processes, after being generated in upstream treatment systems and sewers, and can be readily stripped out and released. Consequently, the current NGERS CH_4 model may need to be modified to factor in the possibility that CH_4 gas stripping can take place on a constant basis within an aerobic treatment system.

Figure 16.3b show that the NGERS N_2O emissions model (Equation 16.4) can make approximate estimations of the actual total N_2O output from an aeration tank operating within a decentralised system. However, the accuracy of these emissions calculations was found to vary significantly from site to site. When compared directly to the measured N_2O emissions value, the NGERS N_2O emissions model had an overall percentage error $(|N_2O_{MODELLED} - N_2O_{MEASURED}|/N_2O_{MEASURED}) \times 100\%)$ of 799% for Site A and a percentage error of 45% for Site C. No percentage error estimate could be calculated for Site B, as N_2O emissions were not released there. However, the NGERS model did estimate that a relatively small amount of N_2O gas (0.2 g $N_2O/m^2/day$) could emit from Site B. These results indicate that the input variables employed by the NGERS N_2O emissions model may not provide enough detail to reliably estimate real-world N_2O fugitive emissions within an acceptable error limit. As such it is appears that the NGERS N_2O emissions model could be redeveloped to factor in site-specific information relating to other important process parameters and operational conditions influencing N_2O generation. These parameters and conditions could include dissolved oxygen level, aeration regime type (intermittent or continuous) and solids retention time. In addition to this, the default N_2O emission factor for domestic wastewater treatment may need to be recalculated and defined specifically for different types of treatment systems running dissimilar treatment processes.

The respective CH_4 and N_2O emission factors calculated over the spring measurement campaign for the Site A, Site B and Site C aeration tanks are provided in Tables 16.3 and 16.4. All of these emission factors were inside the expected range of emission factors that have previously been reported for large-scale centralised activated sludge WWTPs using in-situ gas flux measurement methods (Global Water Research Commission, 2011). The N_2O emission factors calculated for Site A, Site B and Site C are within the expected range of 0% to 4% conversion of influent total N to N_2O emission (Kampschreur *et al.* 2009). Also, the CH_4 emission factors calculated for Site A, Site B and Site C are all within range of CH_4 emission factors estimated for large-scale WWTPs located in the Netherlands and France (0.005 to 0.04) (Global Water Research Commission, 2011). It is possible that these emission factors may increase at each of the sites if CH_4 and N_2O emissions occurring from treatment processes adjoining the aeration tanks are taken into account. However, the overall impact of these adjoining treatment processes may be regarded as being low or negligible, as at each of the decentralised sites the aeration tank systems had the largest spatial footprint and operational volume in comparison to the rest of treatment processes at each particular site and were completely open to the atmosphere. They therefore had the most potential to release the largest amounts of fugitive emissions.

Table 16.3 CH_4 emission factors and the daily average CH_4 emission per m^3 of influent calculated for Site A, Site B and Site C.

Site Name	CH_4 Emission Factor (CH_4EF)	Daily average CH_4 emission per m^3 (g $CH_4/m^2/m^3$)
Site C	0.0088	8.3×10^{-4}
Site B	0.00596	1.1×10^{-3}
Site A	0.0365	1.8×10^{-4}

The daily average CH_4 and N_2O emissions per m^3 of influent calculated over the measurement campaign for Site A, Site B and Site C are given in Tables 16.3 and 16.4. Site B measured the largest daily average CH_4 emission per m^3 of 1.1×10^{-3} g $CH_4/m^2/m^3$, which was an order of magnitude greater than the next highest measurement made for Site C (8.3×10^{-4} g $CH_4/m^2/m^3$). For N_2O, Site C delivered the greatest daily average emission per m^3 of 2.6×10^{-3}g $N_2O/m^2/m^3$, with Site A providing the second highest daily average emission per m^3 of 8.2×10^{-5} g $N_2O/m^2/m^3$. These results indicate that the CH_4 and N_2O output per m^3 of infuent can be relatively high and may be comparable to centralised systems, even for very small decentralised systems like Site B treating small catchment areas.

Table 16.4 N_2O emission factors and the daily average N_2O emission per m^3 of influent calculated for Site A, Site B and Site C.

Site Name	N_2O Emission Factor (N_2OEF)	Daily Average N_2O Emission per m^3 (g $N_2O/m^2/m^3$)
Site C	0.23	2.6×10^{-3}
Site B	0	0
Site A	0.13	8.2×10^{-5}

16.4.2 Total carbon footprint for each reuse system

To evaluate the total daily carbon footprint for each reuse system, the average daily CH_4 and N_2O fluxes measured over the entirety of the measurement campaign reported in Section 16.4.1 were converted to their equivalent CO_2 (CO_{2EQ}) values by multiplying them by their respective 100 year global warming potential conversion factors (25 for CH_4 and 298 for N_2O) given by the Intergovernmental Panel on Climate Change (2007). The average daily CH_4 flux after conversion to the equivalent CO_2 value was calculated to be 50.4 ± 71.4 g CO_{2EQ}/m^2/day for Site A, 1.43 ± 0.46 g CO_{2EQ}/m^2/day for Site B and 4.7 ± 3.3 g CO_{2EQ}/m^2/day for Site C. The daily average N_2O flux following conversion to its equivalent CO_2 value was 336.5 ± 249.9 g CO_{2EQ}/m^2/day for Site A and 220.7 ± 76.6 g CO_{2EQ}/m^2/day for Site C. The total annual carbon footprint due to CH_4 and N_2O emissions for each reuse system was estimated by integrating the average daily CO_{2EQ} CH_4 and N_2O fluxes over the atmospherically exposed surface area of each of the specific treatment processes under analysis and by multiplying this value by the number of days in a common year (365). As a result, the total annual carbon footprint due to CH_4 emissions was 6.8 tonnes (Site A), 0.031 tonnes (Site B) and 0.16 tonnes (Site C). The total annual carbon footprint due to N_2O emissions for each reuse system was calculated to be 45.4 tonnes (Site A) and 7.7 tonnes (Site C).

From these results it is clear that each of the reuse systems emitted relatively significant amounts of CH_4 and N_2O gas (in particular Site A and Site C), which may have an eventual long-term impact upon the atmospheric infrared radiation budget and as a consequence, global climate change. Subsequently, it is important to evaluate various strategies to mitigate and reduce these emissions before they can enter the atmosphere, or to capture and recycle them for other applications, such as providing a localised energy supply to power daily treatment systems operations. This is of critical importance, as the installation of decentralised reuse systems is increasing rapidly in both developed and developing countries, and as such, the cumulative fugitive emissions released from of all these decentralised sites may become greater than the cumulative fugitive emissions output of established centralised treatment systems. Various mitigation and recycling schemes are further detailed in Section 16.4.3.

16.4.3 Emissions mitigation and gas reuse strategies

Strategies for reducing emissions from wastewater treatment processes can have direct and indirect benefits in terms of energy usage and environmental footprint mitigation. The immediate cost/emission avoidance is realised from the generation and exploitation of energy from the capture and reuse of biogas, as well as an indirect reduction of GHG emissions through improved energy and resource efficiency, and a decrease in the use of fossil fuels (Bogner *et al.* 2008). There are many approaches to capturing biogas including covering anaerobic lagoons, membrane capture systems and up flow anaerobic digesters. According to the Australian Water Recycling Centre of Excellence (2010), the anaerobic digester is the most important component of a wastewater treatment process as it generates a constant energy source in the form of biogas, readily mobilises nutrients and it is usually fully enclosed. Biogas capture approaches from anaerobic digesters are usually applied to those treating high strength wastewaters, such as those derived from industrial and agricultural applications.

Biological nitrogen removal is one of the main drivers for treating wastewater and inefficiency in treatment processes and reaction kinetics can lead to the production of high levels of N_2O and other unwanted compounds. Ammonia removal in traditional wastewater treatment processes requires an oxidation phase and a reduction phase and includes multiple process steps. In the denitrification step a carbon source is required if the reaction is to proceed. This is rate limiting and needs careful management. Approaches that utilise anaerobic ammonia oxidation techniques, such as DEMON (DEAmMONification) and ANAMMOX (ANaerobic AMMmonium OXidation), have been developed and provide significant energy and cost saving benefits. An example of a wastewater treatment plant that has utilised anaerobic ammonium oxidation techniques (DEMON) at full scale is the Strass WWTP in Austria. This plant serves a population which varies from 60,000 in summer to 250,000 in winter. In 2004 the plant implemented the DEMON process providing deammonification without the need for a supplemental carbon supply (Wett, 2007). The two specific advantages of employing this process on site were that the energy requirements for nitrification of the side stream ammonia were reduced and the organic sludge previously needed for denitrification of the side stream was now available for conversion to biogas in the anaerobic digesters (WERF, 2010). Following the commissioning of the DEMON process, the onsite specific energy demand of the side-stream process was reduced to 1.16 kWh/kg ammonia nitrogen removed, compared to approximately 6.5 kWh/kg ammonia nitrogen removed using traditional biological nitrogen removal methods at the same plant (Wett, 2007).

Real-world studies, such as the ones described in this chapter, provide a better understanding of the seasonal and operational variability of wastewater treatment processes and associated fugitive GHG emissions. Further and more detailed understanding could be gained via continuous ongoing online real-time monitoring of N_2O and CH_4 gas concentrations as well as process gas concentrations (DO and NH_3), which can be correlated to process conditions and variability in treatment loading. The incorporation of biogas recovery systems, such as a fixed film anaerobic digester or membrane based gas recovery system, to decentralised systems will be site specific and depend on available side streams and existing processes. As additional carbon sources may be required to maximise energy recovery from waste streams, co-location of decentralised wastewater treatment water reuse facilities near other industries that produce high strength wastewaters is highly desirable. A detailed analysis also needs to be performed to determine the long-term cost benefit of deploying such technology at decentralised reuse systems. Additionally, to make a significant impact on energy reduction and resource recovery a step change in current practice is required. Innovative solutions (e.g., the incorporation of technologies such as DEMON or ANAMMOX) for side stream treatment to efficiently remove nitrogen and more effectively utilise the carbon in waste streams for energy recovery, offer considerable potential.

16.5 CONCLUSION

This chapter has described the results of a measurement campaign designed to ascertain levels of CH_4 and N_2O from water reuse facilities at three sites in Melbourne, Australia. It was found that typical decentralised reuse systems running an activated sludge treatment regime can emit greatly variable, but still measurably high levels of CH_4 and N_2O per unit area from their aeration tanks. As decentralised reuse systems are being installed in growing numbers around the world, the cumulative fugitive emissions released from separate decentralised reuse systems serving different communities may become greater than the total fugitive emissions output of established centralised treatment systems. In addition, the emissions data measured at each of the decentralised reuse systems was generally not well correlated to the emissions data calculated by the current NGERS semi-empirical models. Therefore, it is recommended that further revisions be made to these models to make them more applicable to a wider variety of wastewater treatment systems. This will help to greatly improve the accuracy of emissions reporting performed by wastewater treatment operators.

The measured data also showed that both N_2O and CH_4 fluxes can vary dramatically in magnitude over small distances across the surface area of a treatment reactor. Accordingly, neglecting the fact that the magnitude of both N_2O and CH_4 fluxes can vary by measurable amounts over small distances across an aeration tank may result in inaccuracies in recorded N_2O and CH_4 emissions data, and consequently in the overall environmental footprint calculated for the reuse system under investigation. Therefore, in order to gain a more accurate determination of both temporal and spatial flux output, it is recommended that flux measurements are made in rapid succession at multiple evenly spaced positions across the entire measurable surface of an aeration tank.

The emissions dataset presented in this investigation was obtained over a single season. However, modifications to physical and chemical parameters brought on by seasonal change may influence the amount of both CH_4 and N_2O emissions from wastewater treatment processes, particularly variations in sewage temperature, which can have a direct impact upon the production and release of CH_4. Consequently, further field studies spanning across an entire calendar year are required to measure long-term CH_4 and N_2O emissions, in order to better ascertain the overall effect of seasonal change and to assimilate a larger emissions dataset to compare directly to emissions data estimated by the NGERS models. In addition, a direct comparison between the annual emissions output from the decentralised reuse systems and nearby centralised systems treating similar influent is required to quantitatively determine if the decentralised reuse systems produce emissions at a similar magnitude (per litre of influent) to the centralised systems. At this stage, it is not possible to directly compare the emissions data measured from the decentralised reuse systems to any emissions data from centralised systems, as there are no long-term studies available detailing online measurements of CH_4 and/or N_2O gases from centralised wastewater treatment processes in Australia. It is anticipated that this deficiency in the knowledge base will soon be resolved, as a large number of Australian research groups and system operators are beginning to actively measure and record gas emissions from a wide variety of treatment systems.

Although a number of technologies have been developed and implemented, their long term performance in reducing GHG emissions, suitability for different contexts and overall sustainability requires further detailed research.

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