Greenhouse gas emissions from on-site wastewater treatment systems

Submitted to the University of Dublin, Trinity College
for the degree of
Doctor of Philosophy (PhD)

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May 2019
DECLARATION

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Dublin, May 16, 2019

Célia Somlai
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SUMMARY

Domestic wastewater is rich in carbon and nitrogen and given the biogeochemical transformations that these compounds undergo during different treatment processes can be significant sources of greenhouse gas (GHG) emissions. However, most research focus in relation to GHG emissions and wastewater treatment has been on large-scale treatment units with only a small number of studies carried out directly on domestic on-site wastewater treatment systems (DWWTSs) in the field. In the Republic of Ireland, around 500,000 dwellings rely on on-site treatment and disposal of their wastewater, 87% of which use septic tanks (STs) followed by some form of soil treatment unit (STU). Considering the large number of DWWTSs in Ireland, as well more internationally, septic systems are potential significant sources of GHG emissions.

The main aim of this research, therefore, was to develop a better understanding of the contribution of the DWWTSs to the GHG inventories. Four DWWTSs were investigated during this study. One site had a simple one chamber ST with a soakaway, which is common of many older systems in existence, Site 1. Two sites were recently constructed following the latest Irish EPA standards including a two-chamber ST, and/or a packaged secondary treatment unit discharging effluent into an engineered STU, Site 2 and 3. The fourth site had a two-chamber ST followed by a willow-based evapotranspiration system, Site 4. In the STs as well as over the STUs and evapotranspiration systems, discrete and long-term CO$_2$ and CH$_4$ flux measurements were carried out using automated flux chambers and a closed-transient measurement approach. In addition discrete samples were taken for laboratory analysis of N$_2$O.

The soil of the soakaway (Site 1) consumed overall 0.03 kg CO$_2$Eq. yr$^{-1}$ less CH$_4$ and emitted 7.3 kg CO$_2$ yr$^{-1}$ more CO$_2$ than a similarly sized area of control soil. The net GHG emissions were compared from different stages (i.e. ST and STU) of two recently constructed DWWTSs as well as how inclusion of up-front packaged secondary treatment units impact on the net emissions from STU (Site 2 and 3). The total net emissions from the systems were 17.0 and 21.9 kg CO$_2$Eq. cap$^{-1}$ yr$^{-1}$ at the two different sites, respectively. Over 80% of the total net emissions was in the form of CO$_2$, around 15% in CH$_4$ and less than 2% in N$_2$O. GHG fluxes had strong spatial and seasonal variation from the willow bed, Site 4.
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<td>BAF</td>
<td>biological aerated filter</td>
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<td>biological oxygen demand</td>
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<td>CC</td>
<td>Crecora</td>
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<tr>
<td>CH₃COO⁻</td>
<td>acetate</td>
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<td>FTIR</td>
<td>Fourier transform infrared spectroscopy</td>
</tr>
<tr>
<td>GC</td>
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<tr>
<td>GC-ECD</td>
<td>gas chromatography with electron capture detector</td>
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<tr>
<td>GC-FID</td>
<td>gas chromatography with flame ionisation detector</td>
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<tr>
<td>GC-TCD</td>
<td>gas chromatography with thermal conductivity detector</td>
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<td>GHG</td>
<td>greenhouse gas</td>
</tr>
<tr>
<td>GPP</td>
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<td>GWP</td>
<td>global warming potential</td>
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<tr>
<td>H₂O</td>
<td>water</td>
</tr>
<tr>
<td>ICOS</td>
<td>integrated-cavity output spectroscopy</td>
</tr>
<tr>
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<td>infrared</td>
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<tr>
<td>KB</td>
<td>Kilbeggan</td>
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<td>Kilmallock</td>
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<td>LO</td>
<td>Louth</td>
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<td>LTAR</td>
<td>long-term acceptance rate</td>
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<td>MPN</td>
<td>most probable numbers</td>
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<td>nitrogen</td>
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<td>NCP</td>
<td>net carbon exchange</td>
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<td>non-dispersive infrared</td>
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<td>non-dispersive infrared gas analyser</td>
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<td>oxidation reduction potential</td>
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<td>parts per billion</td>
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<td>phosphate</td>
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<td>RBC</td>
<td>rotating biological contractor</td>
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<td>sulphate</td>
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<td>temperature</td>
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<td>total carbon</td>
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<td>total nitrogen</td>
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<td>total oxidised nitrogen</td>
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<td>UASB</td>
<td>uplow anaerobic sludge blanket</td>
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<tr>
<td>UGGA</td>
<td>ultraportable greenhouse gas analyser</td>
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<td>UV</td>
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<td>WWTP</td>
<td>wastewater treatment plant</td>
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1

Introduction

1.1 Background and motivations

The increasing concentrations of greenhouse gases (GHG) in the atmosphere due to emissions from natural and anthropogenic sources are linked to global climate change (IPCC, 2013). In order to mitigate GHG emissions from human derived sources, accurate inventories of the sources and sinks first need to be compiled.

Domestic wastewater is rich in carbon and nitrogen species and given the biogeochemical transformations these compounds undergo during treatment, domestic wastewater treatment processes can be significant sources of GHG emissions. While large-scale, centralised treatment plants have been intensively monitored for their emissions, domestic on-site wastewater treatment systems (DWWTSs) have received little attention when accounting for the release of GHGs.

More than one third of the population of the Republic of Ireland relies on DWWTSs to treat their wastewater (CSO, 2011). The DWWTS most commonly consist of a septic tank (ST) and a soil treatment unit (STU) for effluent dispersal. STs are designed to separate the liquid from the solid fraction of the influent. Settleable solids are retained as sludge at the tank bottom while floatables form a scum layer on the water surface (Gill et al., 2007). By microbial mediated anaerobic processes, the influent organic matter in septic systems is converted into carbon dioxide (CO$_2$) and methane (CH$_4$). The effluent from the tanks is released into the soil for further treatment in the unsaturated zone where potentially additional CO$_2$ and CH$_4$ are produced, as well as nitrous oxide (N$_2$O) from the partial denitrification of nitrate, which then may get emitted to the atmosphere back through the soil.
Considering the large number of DWWTS in the Republic of Ireland, as well as worldwide, septic systems are potential significant sources of GHG emissions nationally and internationally. According to the Irish national GHG inventory for the year 2016, GHG emissions from wastewater treatment and discharge sub-category (combined domestic and industrial) accounted for 147.12 kt CO$_2$Eq., which equates to 15.4% of total emissions from the waste sector and 0.24% of the total national GHG emissions (EPA, 2018a). The existing inventories of GHG emissions from septic systems are based on the assumption of the conversion rate of organic matter entering the systems from a limited number of direct field measurements and only assuming emissions are generated in the STs, considering STUs as negligible sources (IPCC, 2006; USEPA, 2016). There are only two recent studies with direct field measurements of GHG emissions from septic systems; one measuring emissions from STs and the other one from STUs. Diaz-Valbuena et al. (2011) estimated an average 33 g CO$_2$ cap$^{-1}$ d$^{-1}$, 11 g CH$_4$ cap$^{-1}$ d$^{-1}$ and 0.005 g N$_2$O cap$^{-1}$ d$^{-1}$ using static gas flux chambers over the wastewater of eight STs in California, US. In the other study, Truhlar et al. (2016) measured also using chamber method an average 130 g CO$_2$ cap$^{-1}$ d$^{-1}$, $-0.004$ g CH$_4$ cap$^{-1}$ d$^{-1}$ and $0.022$ g N$_2$O cap$^{-1}$ d$^{-1}$ from the STUs of seven septic systems in New York state, US. However, these two studies only include discrete measurements over a short period of time.

1.2 Aims and objectives

The overall aim of this research was to develop a better understanding of the contribution of the DWWTSs to the GHG inventories.

This was carried out via the following targeted objectives:

- Quantifying the on-site production of GHGs (CO$_2$, CH$_4$ and N$_2$O) within DWWTSs using established field research sites set up by the Department of Civil, Structural and Environmental Engineering at TCD.
- Quantifying the GHG emission rates from STs.
- Quantifying the spatial and temporal GHG emission rates from a ST soakaway.
- Quantifying the spatial and temporal GHG emission rates from engineered STUs receiving different levels of wastewater pre-treatment; primary treated ST effluent vs. secondary treated effluents (from a coconut/peat filter and a rotating biological contactor).
• Quantifying the GHG emission rates from a willow-based evapotranspiration system used for on-site wastewater treatment.

• Quantifying the GHG emission production (in CO_{2Eq.}) per person from different DWWTSs in the Irish northern maritime climate.

1.3 Thesis organisation

Chapter 2 presents a comprehensive literature review to outline the current knowledge as well as knowledge gaps, which led to the thesis research objectives.

Chapter 3 describes the field site construction and instrumentation.

Chapter 4 describes the analytical methods.

Chapter 5 investigates CO_{2} emissions from a septic tank soakaway in a northern maritime climate.

Chapter 6 investigates the spatial and temporal variation of CO_{2} and CH_{4} emissions from a septic tank soakaway.

Chapter 7 investigates the GHG emission rates from different stages and levels of wastewater treatments.

Chapter 8 investigates the GHG emissions from an evapotranspiration system.

Chapter 9 summarises the main thesis conclusions and discusses recommendations for future research.
1. INTRODUCTION
2

Literature Review

This chapter presents a comprehensive literature review to outline the current knowledge as well as knowledge gaps. This chapter is divided into five parts.

Section 2.1 presents a brief introduction into DWWTSs and the significance of these systems globally and in the Republic of Ireland. Section 2.2 gives an overview on the different on-site domestic wastewater treatment options including STs, packaged treatments, vegetated systems and soil treatments. Section 2.3 introduces GHGs in general, including CO₂, CH₄ and N₂O. Section 2.4 provides an overall summary of the ongoing biochemical processes through the treatment processes that lead to the production of GHGs in septic systems. This part also includes an overview of previous research studies on GHG emissions from wastewater treatment systems. Section 2.5 gives a comprehensive review of employed methodology on measuring aquatic and terrestrial GHG emissions.
2.1 On-site domestic wastewater treatment

This PhD research focused on on-site domestic wastewater treatment systems (DWWTSs) for individual households. These systems are designed to be cost effective, robust, compact, odourless, requiring little maintenance and protecting public health and water quality (USEPA, 2002).

The following sections will give a short summary on different wastewater management layouts; typical characterisation of domestic wastewater and significance of the on-site domestic wastewater treatment globally and in Europe.

**On-site wastewater treatment** We can distinguish between centralised and decentralised wastewater management as shown in Figure 2.1. Decentralised systems can take the form of small clustered networks down to individual on-site systems.

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**Figure 2.1:** Centralised, decentralised clustered and on-site wastewater management layouts.

Centralised wastewater management consists of a centralised collection system also known as sewer that collects wastewater and transports it to a main centralised wastewater treatment plant on an off-site location outside the settlement, and disposal or reuse of the treated effluent, usually far from the point of origin. During decentralised wastewater management, wastewater is collected, treated and disposed or reused near the point of generation usually from a cluster of homes or a small portion of large communities. On-
2.1. On-site Domestic Wastewater Treatment

Site wastewater management collects, treats and disposes the wastewater at the point of generation (Crites and Technobanoglous, 1998).

The most commonly used on-site systems consists of a septic tank (ST) followed by a soil treatment unit (STU) often known as a percolation area. Some systems also can incorporate a secondary treatment stage before discharge to the soil, in the form of small complete packaged aerated treatment systems, such as rotating biological contractor systems or more passive treatment systems such as constructed wetlands.

**Domestic wastewater** According to the source, wastewater can be categorised as (i) domestic wastewater, also known as sanitary wastewater or sewage, (ii) industrial or trade wastewater and (iii) municipal wastewater, which is the mixture of the two (Gray et al., 2010). The focus of this research, as it was mentioned earlier, is on wastewater from domestic sanitary appliances such as toilets, showers, sinks, wash hand basins, washing machines and dishwashers (EPA, 2013).

Domestic wastewater is 99.9% water by volume and the remaining solid material that requires to be removed is 70% of organic and 30% of inorganic material. The organic fraction is solid material of faeces, food particles, oils, grease, soap and detergents. This fraction is composed primarily of proteins (65%), carbohydrates (25%) and fats (10%). The inorganic part is made of sand, grit and metals (Gray et al., 2010). The strength and composition of sewage varies depending on the design of the system, the volume of wastewater being produced, number of inhabitants, infiltration of groundwater, surface run-off as well as habits of inhabitants (EPA, 2013; Gray et al., 2010).

2.1.1 Contaminants

Domestic wastewater contains a board range of pollutants: microorganisms such as pathogenic bacteria, viruses, and worm eggs are a public health issue, biodegradable organic materials mainly from excreta and kitchen waste can cause oxygen depletion, and macro nutrients (i.e. nitrogen species and phosphorus) can cause ecological problem to surface waters in the form of eutrophication. Other organic compounds from fats, solvents, and detergents contribute to the overall pollution load. Emerging contaminants including endocrine disrupting compounds, personal care products and nanoparticles are increasingly the focus of contemporary research studies. Hence in order to protect public health and the environment, domestic wastewater must be treated to remove these contaminants before being discharged (EPA, 2013).
2.1.2 Significance of on-site domestic wastewater treatment globally and in Europe

Most commonly, DWWTSs are used in more rural areas where the population is low and centralised systems are not cost effective. However, it is currently estimated that 64% of the urban population in low- and middle-income countries use on-site systems as form of sanitation (Hawkins et al., 2014) whilst an additional estimated total of 2.4 billion people are still lacking access to basic sanitation services globally (UNICEF/WHO, 2015). In the United States and in Australia for example, one fifth of the population relies on on-site septic systems (Beal et al., 2005; USEPA, 2016).

In the European Economic Area, the Urban Waste Water Treatment Directive (Communities, 1991) does not enforce policies for agglomerations with less than 2000 population equivalent (PE). However, in Central and Eastern Europe, 42 million people (30% of the population) live in areas where the EU Urban Waste Water Treatment Directive does not apply, of which just 9% are connected to a centralised wastewater treatment plants (Istenic et al., 2015). No data is available on wastewater treatment for the remaining 91%. According to the European Environmental Agency, a total of 23% of European households are estimated to be connected to septic systems (EEA, 2013).

The EN:12566 is a set of European standards which specify the general requirements for on-site wastewater treatment plants used for domestic wastewater treatment for up to 50 PE. It is divided into seven parts; five of them specifying treatment requirements - prefabricated septic tanks (CEN, 2000), packaged and/or site assembled domestic wastewater treatment plants (CEN, 2005b), septic tanks assembled on site from prefabricated kits (CEN, 2007), prefabricated treatment units used for septic tank effluent (CEN, 2013a) and prefabricated tertiary treatment (CEN, 2013b). The two other parts refer to soil infiltration systems (CEN, 2005) and pre-treated effluent filtration systems (CEN, 2008). EN:12566 European standards are implemented in Ireland as SR66 - Guidance on the selection, installation and use of small wastewater treatment systems for domestic wastewater up to 50 PE (NSAI, 2015) as it outlines the requirements for plants to be suitable for use in Ireland. It must be used in conjunction with the Code of Practice entitled Wastewater Treatment and Disposal Systems Serving Single Houses (EPA, 2009).

In the Republic of Ireland, nearly 30% of the households (approximately 500,000 households) rely on individual domestic on-site wastewater treatment systems (CSO, 2011; EPA, 2013), as shown in Figure 2.2. 87% of those households use conventional septic tank
2.1. ON-SITE DOMESTIC WASTEWATER TREATMENT

treatment system. In the Republic of Ireland, ST systems installed before 1991 discharge their effluent released into the soil mainly via a soakaway or soakpit. A soakaway is a large pit back-filled with stone or rubble for effluent disposal and fed from the ST by single effluent pipe. In 1991, the National Standards Authority of Ireland specified that new ST systems should discharge their effluent over a wider percolation areas where the effluent would be distributed through percolation trenches (also known as a leach fields, drainfields, infiltration areas or STUs) (NSAI, 1991). At present, it is estimated that about 65% of ST systems in Ireland were constructed before 1991 (CSO, 2011).

Figure 2.2: Spatial distribution of DWWTSs and sewered areas in Republic of Ireland. Burgundy dots mark individual DWWTSs, blue areas mark locations with access to a centralised sewage network, credit: Jan Knappe.
Improved guidance on the various aspects associated with these DWWTS systems within Ireland is now provided by the Code of Practice for Wastewater Treatment Systems for Single Houses (EPA, 2009), which conforms with EN:12566 and is brought into Irish legislation via Part H of Building Regulations. The EPA has also developed a risk assessment methodology based on density of systems, attenuation and infiltration to establish zones of risk across the entire country to identify where percolation is inadequate (see Figure 2.3a) and where the groundwater vulnerability is high (see Figure 2.3b) which has been used as the basis of the National Inspection Plan (EPA, 2013).

![Figure 2.3](image)

Figure 2.3: (a) Spatial distribution of inadequate percolation areas in the Republic of Ireland and (b) Potential risk to groundwater supplies from DWWTS. Source (EPA, 2018b).

### 2.2 Treatment options

The following sections will give a short introduction to the different DWWTS options; septic tanks, packaged secondary treatment units, vegetated system and final soil treatment.

**Treatment levels** Wastewater treatment systems can be divided into different generic treatment levels, classified as (i) primary, (ii) secondary, and (iii) tertiary treatment. Pri-
mary treatment is a largely physical process which removes larger suspended solids (including organic materials) from the wastewater by settlement. The process also provides a long retention time as well as limited anaerobic decomposition in the case of the on-site treatment system. Secondary treatment is a combination of aerobic microbiological processes which act to decompose the organic matter by microorganisms, for example activated sludge reactors, aerobic stabilisation ponds, trickling filters and anaerobic reactors. Tertiary treatment processes are used to further purify (or "polish") the wastewater of pathogens, contaminants, and remaining nutrients such as nitrogen and phosphorus compounds. In centralised wastewater treatment plants, these processes can include advanced filtration, carbon adsorption, ion exchange, and disinfection (EPA, 2013; Crites and Technobanogloous, 1998). However, for on-site systems much of this treatment is normally achieved within the STU. In certain cases some additional packaged treatment step can be applied to target specific contaminations such as P-removal adsorption filters, or UV disinfection for pathogens.

2.2.1 Septic tank

The most common DWWTS, the septic tank, serves as a combined settling and skimming tank, as an unheated, unmixed anaerobic digester and as a sludge storage (EPA, 2009). The discharge of effluent is released into the soil via a STU (as shown in Figure 2.4) which provides the majority of the treatment through natural physical, chemical and biological processes within the plant-soil-water matrix (Crites and Technobanogloous, 1998).

The ST is one of the oldest units available for the primary treatment of wastewater. STs were first reported in the 1860s in France. In 1881, Abbe Moigno and Louis M. Mouras patented the Fosse Mouras automatic scavenger (Dunbar, 1908; Winneberger, 1984) and their design has remained almost unchanged until today (Crites and Technobanogloous, 1998; USEPA, 2002).
2. LITERATURE REVIEW

Figure 2.4: (a) Plan view of conventional ST and STU layout, source: (EPA, 2009) and (b) longitudinal cross section of typical two-chamber ST with STU.
2.2. TREATMENT OPTIONS

The ST is a buried, watertight, prefabricated tank designed and constructed to facilitate the initial collection and storage of the raw sewage from one or several households. The raw sewage is generally discharged by gravity into the ST. STs can be made of concrete, fiberglass or polyethylene and consist of single or multiple chambers. The most common configuration is rectangular with interior baffles to divide the tank and to access ports for maintenance (Gray et al., 2010; Crites and Technobanogloous, 1998).

There are three characteristic layers in the ST: the sludge layer at the bottom, scum at the top and a clear zone in the middle, see Figure 2.4b. The sludge layer is formed by the retention of settleable particles at the bottom of the tank, where facultative and anaerobic decomposition of organic matter take place, see Section 2.4. The scum layer is composed of accumulated floating materials, as greases, oils, and other buoyant particles. The scum layer is not vital, but it helps the operation by preventing oxygen transfer through the air-water surface, preventing heat loss and retaining any solids that rise from the sludge layer. In regular intervals, scum and sludge have to be removes with a vacuum truck. Gill et al. (2018) found in the Republic of Ireland the recommended desludging interval of once every 3 years. This has to be done by professionals. The clarified effluent is discharged into the soil via an engineered STU. The complex biochemical transformations of the ST are considered in more detail in Section 2.4.1.

2.2.2 Packaged treatment systems

A packaged system can be installed as an alternative to a ST or following the ST to provide further treatment of ST effluent as a secondary (aerobic microbiological) treatment prior to discharge to subsoil. In order to reduce organic matter and level of pathogens, these systems use mechanical parts (either pumps/or aerators) to enhance the aerobic treatment of domestic wastewater and allow the wastewater to come in contact with a dense microbiological community either suspended in a reactor or by providing a large fixed surface area for the biofilm growth. All packaged treatment systems requires regular monitoring and maintenance.

The main types of packaged treatment systems are briefly described below:

Biological aerated filter systems

The biological aerated filter (BAF) systems combine filtration with biological treatment, see Figure 2.5. BAF systems consist of a primary settlement tank, an aerated submerged biofilm filter and a secondary settlement tank. The media of the filter normally have a high
specific surface area and can consist of plastic modules or granular material. This media supports the development a biologically active layer, which contains complex bacterial polysaccharides, accumulated organic substances and microorganisms and this biofilm treat the wastewater. BAF systems are constructed in glass-reinforced plastic, concrete or steel.

Figure 2.5: Schematic of BAF system, source: (EPA, 2009).

**Sequencing batch reactor systems**

The sequencing batch reactor (SBR) process is a form of activated sludge treatment in which aeration, settlement and decanting can occur in a single reactor, see Figure 2.6. The process employs a five-stage cycle: fill, react, settle, empty and rest. Wastewater enters the reactor during the fill stage, the mixing is provided by mechanical means under anoxic condition. Aeration is performed during the second stage by the use of fixed or floating mechanical pumps or by transferring air into fine bubble diffusers fixed to the floor of the tank. No aeration is provided during the third stage and the suspended solids settle down to the bottom of the reactor. During the fourth stage the supernatant is decanted and sludge is withdrawn from the reactor. After the rest stage the cycle commences again with a new fill stage. This required precision means that these SBR systems should have high levels of maintenance and control.
2.2. **TREATMENT OPTIONS**

**Activated Sludge Systems**

Activated sludge systems usually promote the growth of a suspended biological flocculation of organic matter that substantially removes organic material through the addition of oxygen. These systems usually consist of two tanks with the first proving aeration to the effluent and then the second allowing the settlement of the biomass as sludge before discharge of the supernatant to a soil treatment area.

**Membrane filtration systems**

Membrane filtration systems treat effluent by the removal of both suspended solids and dissolved molecular material from the effluent as it passes across a specific membrane material, see Figure 2.7. The system utilise a treatment tank with aeration and membrane filtration units. These systems usually produce very high quality effluents. The special membrane used is mounted on a support frame and in order for the effluent to progress from the inlet end of the system to the outlet end, it should pass through the membrane unit. These systems need to be cleaned regularly and so have high operating costs.
2. LITERATURE REVIEW

Figure 2.7: Schematic of membrane filtration system, source: (EPA, 2009).

Media filter

A media filter is a type of trickling filter that uses a bed of peat, coconut husks, shredded tyres, crushed glass, geo-textile fabric or other medium material to provide a large surface area for the fixed growth of a biofilm across which wastewater passes for treatment, see Figure 2.8. Such filters typically consist of a distribution system, the treatment media and a drain. Septic tank wastewater is intermittently dosed evenly, via a pipe distribution network fitted with orifices onto the top of the media. The effluent then percolates through the media receiving treatment by passive biofiltration processes (filtration, absorption, adsorption, ion exchange, microbial assimilation etc.). Peat as a media filter for example, is polar, has a high surface area and a highly porous structure. In addition, the low pH of the peat media, its trace hydrocarbons and indigenous microflora have some anti-microbial properties.
2.2. TREATMENT OPTIONS

Rotating biological contactor

Rotating biological contactor (RBC) systems consist of a primary settlement tank, a secondary treatment compartment with rotating disks and a secondary settlement tank, see Figure 2.8. The rotating disks provide a media with high surface area where biofilm can develop. This biofilm can break down and stabilize organic pollutants contained within the wastewater in an aerobic environment which is maintained by a rotating disk which continually brings them into contact with the oxygen in the atmosphere as the disk rotates.
2.2.3 Vegetated systems

Vegetated treatment systems are nature–based solutions for domestic wastewater treatment and can be designed to receive either primary or secondary treated effluent. Similarly to natural wetlands, vegetated systems act as a biofilter and are capable of removing a range of pollutants such as organic matter, nutrients, pathogens and heavy metals from the water as it passes through the system (Sasse, 1998; Tilley et al., 2014). Systems commonly used in the Republic of Ireland include constructed wetlands and zero–discharge willow evaporation beds (Gill et al., 2004; Curneen and Gill, 2013, 2016). Both types generally consist of a sealed and backfilled (with either gravel or soil) treatment basin with planted vegetation where the thick root mass acts as a pathway for the transfer of oxygen from the atmosphere to the root zone, the so–called rhizosphere. Due to the sealed treatment basin, these systems can be suitable for areas where the soil infiltration is limited due to the presence of very poor permeability subsoil (Curneen and Gill, 2016). A wide variety of vegetation has been successfully used in vegetated systems, e.g., common reed (*Phragmites australis*), other plants species used are *Iris, Typha, Sparganium, Carex, Schoenoplectus* and *Acorus*; however, optimal choices strongly depend on local climate, soil and effluent characteristics. A mixture of plant species is generally encouraged to promote ecosystem diversification in the system.
2.2. TREATMENT OPTIONS

**Constructed wetlands (reed beds)**

There are three main types of constructed wetlands: (i) surface–flow systems, (ii) subsurface–flow systems and (iii) hybrid systems. In surface–flow or overland systems the water is distributed above ground, mainly using mounted distribution pipes or sprinklers (Sasse, 1998). Effluent will flow above ground and is exposed to the atmosphere and direct sunlight. While this enhances pathogen removal by UV irradiation, the predominantly ponded conditions can be a breeding ground for insects such as mosquito and can pose a direct potential health risk due to partially treated effluent being exposed (Tilley et al., 2014). The soil layer below the water is generally anaerobic, but oxygen may be abundant in the rhizosphere. Subsurface–flow systems can be implemented with either horizontal or vertical flow. Both rely on attached growth microbial organisms coupled with the planted vegetation to facilitate pollutant removal. However, while vertical flow systems generally operate with discrete pulsed flows and corresponding aerobic/anaerobic cycles similar to a trickling filter, horizontal flow wetlands have a permanent (sub–surface) water table, as shown in Figure 2.10. Although facultative and anaerobic bacteria degrade most organic matter, the vegetation transfers a small amount of oxygen into the root zone and aerobic bacteria can colonise the rhizosphere (Tilley et al., 2014). More complex hybrid systems combine different types of constructed wetlands to achieve an improved treatment efficiency by combining the advantages of individual systems – host hybrid systems combine vertical and horizontal subsurface–flow stages and offer the potential for denitrification if effluent is recirculated (Tilley et al., 2014).

![Figure 2.10: Schematic of horizontal subsurface flow reed bed, source: (EPA, 2009).](image-url)
Evapotranspiration (willow) systems are mostly used in the Republic of Ireland and Denmark. Willow belongs to the genus *Salix* and family *Salicaceae* and is a moisture loving plant. Willow has numerous physiological traits which make it highly suitable for phytoremediation purposes. Firstly, willow has high growth (mean crop growth between 6 and 18 g m\(^{-2}\) d\(^{-1}\) in a Scottish study by (Cannell et al., 1987) and productivity (dry matter production was reported up to 30 tonnes ha\(^{-1}\) yr\(^{-1}\) in Sweden by (Christersson, 1986)) and a relatively high capacity among woody plants to convert solar radiation into chemical energy under certain climatic conditions (growth efficiency ranged between 0.99 and 1.38 g MJ\(^{-1}\) by (Cannell et al., 1987)). Secondly, many willow species have an extensive fibrous root system, with, approximately 80% of the fine root hairs found at depths of less than 40 cm and a continuous growth of the fine roots for a large proportion of the growing season from May until September (Rytter and Hansson, 1996). Thirdly, it can achieve high evapotranspiration rates throughout the growing season (with the mean evapotranspiration during growing season of 2.6 mm d\(^{-1}\) measured both in Sweden by (Persson and Lindroth, 1994) and in the Republic of Ireland by (Curneen and Gill, 2016)), which is a combination of removal of water from the soil surface to the atmosphere via a combination of evaporation, direct movement of the water from the soil; and transpiration, movement of water within the plant. Fourthly, willow has been proved to have efficient uptake of nutrient compounds (Ericsson, 1981; Ellsworth, 1999; Dimitriou and Aronsson, 2011) (31% of the applied fertilizer N was accounted for (Hangs et al., 2012)); high filtering capacity for nitrogen and phosphorus (N and P retention was up to 96% and 94%, respectively (Dimitriou and Aronsson, 2011)); ability to facilitate denitrification in the root zone (Aronsson and Bergstrom, 2001); ability to accumulate high levels of toxic metals, especially cadmium (Klang-Westin and Eriksson, 2003; Dickinson and Pulford, 2003). Lastly, willow has a high tolerance for flooded or saturated conditions and oxygen shortage in the root zone (Krasny et al., 1988; Aronsson and Perttu, 2001). Jackson and Attwood (1996) found that willow roots survived at least 4 weeks of waterlogging at depths of at least 300 mm and showed ability to recommence growth rates when the soil was drained.

However, studies in the Irish context found that willow system did not manage to achieve complete zero-discharge in any year but remained at maximum water level for most of the winter months, indicating some loss of water by lateral exfiltration at the surface (Curneen and Gill, 2016). However, chemical and microbiological sampling of water in the sumps and ponded water over the winter periods showed good water quality, equivalent to surface runoff from the adjacent field, and so the systems were acting as good pollutant attenuation devices, even if they could not be described as fully zero-discharge systems across an annual basis. The cross section and top view of the evapotranspiration system used by (Curneen and Gill, 2016) are shown in 3.15a and 3.15b, respectively.
2.2. TREATMENT OPTIONS

Figure 2.11: (a) top view of layout of 110 mm percolation distribution piping and (b) cross section showing two rows of distribution piping in the gravel layer, source: (Curneen and Gill, 2016).

2.2.4 Soil treatment

The final element of the treatment process is discharging treated wastewater into the groundwater via the unsaturated subsoil, which is commonly referred as a percolation area, a leach field or an infiltration area; the preferred term used in this thesis is soil treatment unit (STU). During this final step, the effluent from the ST or packaged treatment system is discharged into the subsoil and undergoes physical straining, chemical ion exchange and adsorption and biological processes before recharge to groundwater.
Biomat is a heterogeneous clogging layer in the subsoil, which is the key component of the treatment process of the STU. There are a number of factors that affect the development of a biomat layer including; the hydraulic loading rate, the dosing regime, the composition of the wastewater, the aeration status of the infiltrative surface and the soil biogeochemical properties (Siegrist and Boyle, 1987; Beal et al., 2006; McKinley and Siegrist, 2011). Beal et al. (2005) has described three phases of biomat development: phase one consists of the initial physical clogging of the pores due to an accumulation of suspended solids, organic matter, and chemical precipitation and can result in reduced infiltration rates over the first few months after installation. Following this initial phase, a period of gradually decreasing infiltration rates takes place and anaerobic biological activity is dominated (Beal et al., 2006). During the final phase, an equilibrium state is being reached, usually with low infiltration rates (Siegrist and Boyle, 1987). This longer residence time of the wastewater in the unsaturated zone results in more effective the removal rates of pathogens and chemicals. According to Gill et al. (2009), the development of a biomat will be significantly retarded at STUs receiving highly treated effluent such as that discharged by a secondary treatment system due to the low organic content. Consequently hydraulic loading rates in percolation areas receiving secondary treated effluent can be higher than those receiving septic tank effluent.

In order to use subsoil for the treatment and disposal of wastewater, the hydraulic assimilation capacity and permeability of the subsoil under saturated conditions must first be determined. Long term acceptance rate (LTAR, \([\text{L m}^{-2} \text{d}^{-1}]\)) is defined as the amount of pre-treated effluent which the system can infiltrate during its lifetime without water logging and clogging (Beal et al., 2006). In the Republic of Ireland, a falling head test is conducted in-situ and calculates the average time (minutes) for water to drop 25 mm at the depth of 400 mm below the invert level of the proposed percolation system pipes. A site is deemed as being acceptable for the installation of a septic tank if the this value is less than 50 or for installation of a secondary treatment system if the value is less than 75.

Traditionally STUs were just simple soakpits, but since 1991 the legislation in the Republic of Ireland has required a much larger percolation area to be constructed comprised of a more complex perforated pipe system located in gravel filled excavated trenches, more details are in the following sections.
2.2. TREATMENT OPTIONS

Soakaway

Soakaways, also referred to as seepage pits or soak-pit systems, are large pits back filled with stone and rubble that are receiving effluent from the ST by a single gravity fed effluent distribution point. Although soakaways can provide a considerable level of treatment within areas of moderate permeability, they pose a high risk of pollution in areas of very high and low permeability subsoil (Keegan et al., 2013). In areas with high permeability subsoil, soakaways may not provide sufficient time for the pollutant attenuation as the effluent percolates rapidly through the vadose zone posing a risk to groundwater quality. In areas with low permeability subsoil, soakaways often fail as the hydraulic load is too great for the subsoil and effluent eventually backs up causing surface ponding with an increase in surface run-off to nearby streams and rivers and risk to human health.

Engineered soil treatment units

An engineered STU is a series of subsurface percolation trenches, see Figure 2.12a and 2.12b. The wastewater is usually allowed to flow by gravity into a distribution box, which distributes the flow evenly into a minimum of four percolation pipes (in the Republic of Ireland) in the percolation trenches. However, in some cases the effluent may need to be pumped up to a higher level before being allowed to flow by gravity through the trenches. The pipes are usually placed over a layer of gravels, which help to distribute the wastewater as well as provide a medium for initial treatment of the effluent. The effluent then percolates into the subsoil, where it undergoes further biological, physical and chemical interactions as described above.
2.3 **Greenhouse gases**

2.3.1 **GHGs and climate change**

GHGs account for less than 0.5% of the volume of the atmosphere, however, they are able to alter Earth’s climate by absorbing energy in the lower atmosphere and re-emitting it, see Figure 2.13. The climate of Earth is controlled by the balance between incoming solar radiation (341 W m⁻² yr⁻¹, global mean) and reflected solar radiation (from clouds and different surfaces, 102 W m⁻² yr⁻¹) together with the emitted thermal infrared radiation (239 W m⁻² yr⁻¹), this balance on average should be zero to sustain the climate. Some of the solar radiation (78 W m⁻² yr⁻¹) and most of the infrared radiation (356 W m⁻² yr⁻¹) emitted at the Earth’s surface are absorbed by GHGs and re-emitted in both upward
(199 W m$^{-2}$ yr$^{-1}$) and downward (as back radiation, 333 W m$^{-2}$ yr$^{-1}$) directions. This process is naturally occurring and crucial to maintain living conditions on the Earth; however, the increase in atmospheric GHGs has a significant impact on Earth’s climate because Earth’s incoming and outgoing radiation is out of balance and forces the climate to change (Trenberth et al., 2009; IPCC, 2013).

Figure 2.13: The global annual mean Earth’s energy budget for March 2000 and May 2004. Radiative forcing [W m$^{-2}$], is any imposed perturbation on the Earth’s energy balance. The broad arrows indicate the schematic flow of energy in proportion to their importance. Source: (Trenberth et al., 2009).

The recent significant changes in our climate system are unprecedented. The pH of ocean surface water has decreased by 0.1 since the beginning of the industrial era, global mean sea level rose by 0.19 over the period 1901 to 2010 and the average rate of ice loss from glaciers around the world was estimated to 275 Gt yr$^{-1}$, naming only few of the major problems, have demonstrated changes in the global water cycle and in the global climate system. The globally averaged combined land and ocean surface temperature shows a warming of 0.85°C over the period 1880 to 2012, see Figure 2.14. In particular, the last three decades have exceeded any preceding decade since 1850.

This increase is cannot be attributed to natural climate fluctuation (IPCC, 2013).

According to the IPCC (2013), the main cause of the climate change is the increasing atmospheric level of GHGs since the Industrial Revolution, see Figure 2.15. This increase of GHGs is caused by anthropogenic emissions from the use of fossil fuel as a source of energy and from land use and land use changes, in particular the intensification of
agriculture. The economic and population growth have been the most important drivers of increases in man-made emissions. In 2010, anthropogenic GHG emissions reached $49 \pm 4.5 \text{ Gt CO}_2\text{Eq. yr}^{-1}$ and half of the anthropogenic CO$_2$ emissions between 1750 and 2010 have occurred in the last 40 years.

Concern about climate change and increasing atmospheric levels of GHGs (see Figure 2.15) has resulted in intensive research on the identification and quantification of all natural and anthropogenic sources and sinks of CO$_2$, CH$_4$ and N$_2$O (IPCC, 2013; USEPA, 2016), with special focus placed on the anthropogenic perturbation of the different pathways of fluxes (Ver et al., 1999; Regnier et al., 2013).

Figure 2.15: Globally averaged atmospheric concentrations of CO$_2$ (green), CH$_4$ (orange) and N$_2$O (red) determined from ice core data (dots) and from direct atmospheric measurements (lines). Source: (IPCC, 2013).
2.3. GREENHOUSE GASES

2.3.2 CO₂, CH₄ and N₂O

The concentrations of CO₂, CH₄ and N₂O have rapidly increased in the atmosphere exceeding the pre-industrial levels by about 40%, 150% and 20%, respectively. These three GHGs altogether amount to 80% of the total radiative forcing from well-mixed GHGs (IPCC, 2013).

<table>
<thead>
<tr>
<th>Characteristics</th>
<th>CO₂</th>
<th>CH₄</th>
<th>N₂O</th>
</tr>
</thead>
<tbody>
<tr>
<td>Molar mass [g mol⁻¹]</td>
<td>44.01</td>
<td>16.04</td>
<td>44.01</td>
</tr>
<tr>
<td>Appearance</td>
<td>Colourless</td>
<td>Colourless</td>
<td>Colourless</td>
</tr>
<tr>
<td>Odour</td>
<td>Odourless</td>
<td>Odourless</td>
<td>Sweetish</td>
</tr>
<tr>
<td>Density at 1 atm and 0°C [kg m⁻²]</td>
<td>1.977</td>
<td>0.717</td>
<td>1.977</td>
</tr>
<tr>
<td>Solubility in water [mg l⁻¹]</td>
<td>1450</td>
<td>22.7</td>
<td>1500</td>
</tr>
<tr>
<td>Atm concentration [ppm]</td>
<td>411</td>
<td>1.86</td>
<td>0.33</td>
</tr>
<tr>
<td>GWPᵃ</td>
<td>1</td>
<td>25</td>
<td>298</td>
</tr>
</tbody>
</table>

ᵃ The global warming potential (GWP) is quantification of the relative radiative forcing impacts of a particular GHG averaged over time (100 yr) and expressed relative to the climate influence of an equivalent mass of CO₂ emission.

CO₂ is a colourless, odourless gas consisting of molecules made up of two oxygen atoms and one carbon atom, see Table 2.1. It is produced when an organic carbon compound (such as wood) or fossilized organic matter (such as coal, oil, or natural gas) is burned in the presence of O₂. CO₂ is taken up through absorption by seawater and photosynthesis by ocean-dwelling plankton and land plants; however, seawater is also a source of CO₂ to the atmosphere, along with land plants, animals, and soils, when CO₂ is released during respiration.

As discussed in earlier in this Section, the level of CO₂ in the atmosphere is increasing due to human activities; primarily fossil fuel combustion (petroleum, coal, natural gas) and secondly land use change. The global atmospheric CO₂ level has increased by an average of 2.2 ppm per year over the last decade (2008-2017) and by almost 15% over the last 250 years, see Figure 2.13. The CO₂ level was 411.2 ppm on 29th June 2018 according to http://www.co2levels.org/.

56% of the CO₂ emitted as human activities is accumulating in the atmosphere and 44% is being taken up by oceans and biosphere. Within the atmosphere, all GHGs except CO₂ and water vapour are removed from the atmosphere primarily by chemical processes. CO₂ is practically inert in the atmosphere and does not directly influence the chemistry. However, atmosphere is an in-situ source of CO₂ from oxidation of CH₄, CO and VOC.
\( \text{CH}_4 \) is a colourless, odourless non-toxic organic trace gas consisting of molecules made up of four hydrogen atoms and one carbon atom, see Table 2.1. \( \text{CH}_4 \) is combustible at concentrations of 5–15\% in air, and it represents the major component of natural gas, biogas, and marsh gas. \( \text{CH}_4 \) is the most abundant organic species in the Earth’s atmosphere and a reactive trace gas arising from both natural and anthropogenic sources. \( \text{CH}_4 \) is released when organic matter decomposes in low oxygen environments, with natural sources including wetlands, swamps and marshes, termites, and oceans. Human-related sources include the mining of fossil fuels and transportation of natural gas, digestive processes in ruminant animals such as cattle, rice paddies and buried waste in landfills. After \( \text{CO}_2 \), \( \text{CH}_4 \) is the second most important GHG as it absorbs long-wave radiation reflected from the Earth’s surface and has an estimated global warming potential per molecule 25 times greater over a 100 year horizon and 72 times greater over a 20 years horizon in comparison to \( \text{CO}_2 \) (IPCC, 2013). However, while being usually oxidised to \( \text{CO}_2 \) and \( \text{H}_2\text{O} \) in upper layers of the atmosphere, its mean atmospheric lifetime is estimated to be approximately 8 – 12 years (Wahlen, 1993). \( \text{CH}_4 \) is removed primarily in the atmosphere by photo-oxidative and hydroxyl radical associated processes. This removal takes place in the troposphere, the lowermost part of the atmosphere.

The global atmospheric \( \text{CH}_4 \) level was 1859.1 ppb on March 2018 according to \url{http://www.methanelevels.org/} and it has been more than doubled in the last 250 years, see Figure 2.13.

\( \text{N}_2\text{O} \) is a colourless, non-flammable gas with a sweetish odour, commonly known as 'laughing gas', and sometimes used as an anaesthetic, see Table 2.1. \( \text{N}_2\text{O} \) is naturally produced in the oceans and in rainforests from incomplete denitrification of nitrate. Anthropogenic sources of \( \text{N}_2\text{O} \) include the use of fertilizers in agriculture, nylon and nitric acid production, cars with catalytic converters and the burning of organic matter. \( \text{N}_2\text{O} \) cannot react with hydroxyl radicals and it is broken down in the atmosphere by chemical reactions driven by sunlight in the stratosphere and above.

\( \text{N}_2\text{O} \) level was 330.9 ppb in May 2018 according to \url{http://www.n2olevels.org/} this is more than 20\% higher than pre-industrial level and it continues to slowly increase over time with an average rate of 0.9 ppb yr\(^{-1}\) and it has a global warming potential 298 times that of \( \text{CO}_2 \) for a 100-year time scale, see Figure 2.13.
2.3.3 Global cycles of CO$_2$, CH$_4$ and N$_2$O

CO$_2$, CH$_4$ and N$_2$O have large, natural emission rates, which have varied over past climatic changes although in more recent history have sustained a stable atmospheric level for centuries prior to the Industrial Revolution, until the recent rapid changes that have occurred since then. The observed change in the atmospheric concentration of CO$_2$, CH$_4$ and N$_2$O results from the dynamic balance between anthropogenic emissions, and the perturbation of natural processes. Natural processes are linked to physical conditions, chemical reactions and biological transformations and they respond themselves to perturbed atmospheric composition and climate change. Therefore, the physical climate system and the biogeochemical cycles of CO$_2$, CH$_4$ and N$_2$O are coupled.

Global cycle of CO$_2$

![Figure 2.16: Simplified schematic of the global carbon cycle. Source: (IPCC, 2013).](image)

The CO$_2$ cycle is, by far, the largest of the three with a total annual turn-over rate of 710 T g yr$^{-1}$ of C between its individual reservoirs and an estimated 10.5% being driven by CO$_2$ releases into the atmosphere through anthropogenic uses, see Figure 2.16.
The black numbers and arrows indicate reservoir mass and exchange fluxes estimated for the time prior to the Industrial Era and the red arrows and numbers indicate annual perturbation of the carbon cycle in fluxes averaged over the 2000-2009 time period. The majority of these anthropogenic CO\textsubscript{2} emissions stems from the burning of fossil fuel and cement production, but also includes agriculture, forestry and other land use changes. It is estimated that all these processes add an additional 4 Pg of C into the atmosphere every year.

Global cycle of CH\textsubscript{4}

![Figure 2.17: Simplified schematic of the global CH\textsubscript{4} cycle. Source: (IPCC, 2013).](image)

The global atmospheric burden of CH\textsubscript{4} is estimated to be approximately 4950 Pg, equivalent to an average concentration of 1885 ppb with an average annual increase of 5.7 ppb mainly due to anthropogenic drivers (Nisbet et al., 2016), see Figure 2.17. According to the IPCC (2013), current CH\textsubscript{4} concentrations in the atmosphere have not been exceeded within the last 420,000 years.

Biogenic CH\textsubscript{4} is produced under anaerobic conditions in usually wet environments like wetlands, bogs, fens, rice field, and landfill sites or in the stomachs of ruminant animals, by
mainly two bacterial pathways: acetate fermentation or CO$_2$ reduction. Landfill sites provide ideal anaerobic environments for methanogenic bacteria breaking down waste organic matter. But also waste handling facilities such as wastewater and sludge treatment processes often facilitate similar conditions of carbon abundance and oxygen depletion which can lead to significant amounts of CH$_4$ being produced. Other, non-biogenic, sources include coal mining activities and biomass burning. While annual production rates and the magnitude of different sources and sinks remains uncertain, estimates suggest that up to 42.8% of the total annual 880 T g yr$^{-1}$ emissions of CH$_4$ to the atmosphere are related to human activity. CH$_4$ produced in anoxic pockets in natural soils can be substantially reduced by bacterial CH$_4$ oxidation (also termed high affinity oxidation) in the soil (Wahlen, 1993); up to 47 P g yr$^{-1}$ of CH$_4$ are estimated to be removed in the upper layers of soil. However, it has been shown that exposure of soils to high levels of ammonium can lead to a loss of methanotrophic bacteria and a subsequent reduction in natural CH$_4$ oxidation, thus, being detrimental to the removal of CH$_4$.

Global cycle of N$_2$O

![Figure 2.18: Simplified schematic of the global N$_2$O cycle. Source: (IPCC, 2013)](image-url)
The globally averaged surface abundance of \( \text{N}_2\text{O} \) is estimated to be approximately 330 ppb, corresponding to a global atmospheric burden of 1530 Tg. Fluxes of \( \text{N}_2\text{O} \) between its global reservoirs all nearly entirely unidirectional, i.e. leading from both the hydrosphere and pedosphere into the atmosphere, see Figure 2.18. Anthropogenic uses contribute nearly 60% to the overall atmospheric fluxes of \( \text{N}_2\text{O} \) and stem mainly from industrial uses, fossil fuel and biomass burning and agriculture, but a significant amount (up to 0.3 T g yr\(^{-1}\) of N) also comes from human excreta and its processing in wastewater treatment.

2.3.4 Biogenic GHG exchange in ecosystems

Exchange of biogenic trace gases (such as \( \text{CO}_2 \), \( \text{CH}_4 \) and \( \text{N}_2\text{O} \)) between surfaces (soil or water) and the atmosphere depend on the production and consumption of the gases by microbial processes, on physical transport through soils, sediments or water and on flux across surface-air boundaries. These biological and physical processes depend on other biotic and abiotic properties of ecosystems, controlling processes (Matson and Harriss, 1993). Water surfaces and soils, depending on the conditions, act as sources or sinks of \( \text{CO}_2 \), \( \text{CH}_4 \) and \( \text{N}_2\text{O} \) (Jungkunst and Fiedler, 2007) and precise qualifications are needed to obtain reliable global budgets. A variety of techniques have been developed to measure surface-atmosphere gas exchange, each method has advantages and disadvantages, and no single approach is applicable to all studies, (see Section 2.5 for more detail).

2.4 GHG emissions from wastewater treatment processes

GHGs can be produced and emitted at many stages between wastewater sources and final disposal (Bogner et al., 2007); direct emissions from the biological processes and indirect emissions resulting from the energy generation. Soluble organic matter (OM) in wastewater is generally removed via biological processes in which microorganisms consume the OM for maintenance and growth under aerobic or anaerobic conditions and produce \( \text{CO}_2 \) and \( \text{CH}_4 \); this production depends on the quantity of degradable OM in the wastewater, the temperature, and the type of treatment system. In addition, \( \text{N}_2\text{O} \) is an intermediate product of microbial N cycling promoted under reduced aeration, high moisture and abundant nitrogen (Kampschreur et al., 2009; Law et al., 2012; USEPA, 2016).
2.4.1 Biogeochemical processes in septic systems

Septic systems rely solely on uncontrolled and naturally occurring biogeochemical processes in order to treat wastewater. [Wilhelm et al. (1994)] presented a conceptual model, which describes this complex biogeochemical evolution of domestic wastewater in conventional on-site septic systems. Two major redox environments were identified - the ST and the STU, as shown on Figure 2.19.

![Conceptual model of the biogeochemical evolution of domestic wastewater in conventional on-site septic systems. Source: Wilhelm et al. (1994).](image)

The first redox zone mainly occurs in the ST, where the concentration of organic C and N is very high and the concentration of the dissolved oxygen (DO) is very low, see Figure 2.20 - Equations: 2.1 - 2.9. In this anaerobic environment, microbiological degradation of organic matter via hydrolysis, acidogenesis, and methanogenesis produces CO$_2$, CH$_4$ and ammonium (NH$_4^+$).

In the STU both anaerobic and aerobic conditions can exist. Gaseous diffusion through the unsaturated sediments of the STU supplies O$_2$ for aerobic oxidation of organic C and NH$_4^+$ forming CO$_2$ and NO$_3^-$ see Figure 2.20 - Equations: 2.10 - 2.11. If there is not adequate O$_2$ the aerobic digestion is incomplete and NO$_3^-$ is reduced to N$_2$ by anaerobic process of denitrification, see Figure 2.20 - Equations: 2.12.
2. LITERATURE REVIEW

### 1st Anaerobic Zone - ST

#### Organic Molecule Hydrolysis

- Proteins + H₂O → Amino Acids \( (2.1) \)
- Carbohydrates + H₂O → Simple Sugars \( (2.2) \)
- Fats + H₂O → Fatty Acids and Glycerol \( (2.3) \)

#### Ammonium Release

- Urea[CO(NH₃⁺)]₂ + H₂O → 2 NH₄⁺ + CO₂ \( (2.4) \)
- Amino Acids + H₂O → NH₄⁺ + Organic Compounds \( (2.5) \)

#### Fermentation

- Amino Acids, Simple Sugars → H₂, CH₃COO⁻, Other Organic Acids \( (2.6) \)

#### Anaerobic Oxidation

- Fatty Acids + H₂O → H₂, CH₃COO⁻ \( (2.7) \)

#### Methanogenesis

- CH₃COO⁻ + H⁺ → CH₄ + CO₂ \( (2.8) \)
- CO₂ + 4 H₂ → CH₄ + 2 H₂O \( (2.9) \)

### Aerobic Zone - STU

#### Organic Matter Oxidation

- Organic Matter + O₂ → CO₂ + H₂O \( (2.10) \)

#### Nitrification

- NH₄⁺ + 2 O₂ → NO₃⁻ + 2 H⁺ + H₂O \( (2.11) \)

### 2nd Anaerobic Zone - STU

#### Denitrification

- 4 NO₃⁻ + 5 CH₂O + 4 H⁺ → 2 N₂ + 5 CO₂ + 7 H₂O \( (2.12) \)

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Figure 2.20: Important biogeochemical reactions in the septic system - adapted from [Wilhelm et al., 1994].

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**GHG emissions from ST**

As mentioned above, the ST provides an environment where anaerobic digestion processes occur, and the organic components of the wastewater are converted to CH₄ and CO₂ by separate groups of microorganisms.
There are four distinct steps; 1 hydrolysis of biopolymers, 2 fermentation of amino acids and sugars, 3 anaerobic oxidation of long chain fatty acids, alcohols and intermediary products such a volatile acids and 4 methanogenesis; that can divided into 2 main stages; (i) waste conversion and (ii) waste stabilisation (McCarty, 1964).

During the **hydrolysis of biopolymers**, hydrolytic bacteria break down lipids, complex polymeric molecules (e.g. protein and carbohydrate) and particulate OM into simpler soluble monomers such as short chain fatty acids, glycerol, peptides, amino acids, oligosaccharides and sugars. These extracellular enzymes (*Clostridium, Peptococcus, Vibrio, Micrococcus* and *Bacillus*) are able to access large substrate molecules.

The monomers produced by the hydrolytic bacteria during the first stage of the anaerobic digestion, go through **fermentation** by many different fermentative genera and species (*Clostridium*, *Bacteroides*, *Ruminococcus*, *Butyribacterium*, *Propionibacterium*, *Eubacterium*, *Lactobacillus*, *Streptococcus*, *Pseudomonas*, *Desulfobacter*, *Micrococcus*, *Bacillus* and *Escherichia*). During fermentation, several intermediate products are produced, namely acetate, propionate, butyrate and hydrogen.

Acid forming bacteria consists of both acidogenic (organic acid forming) and acetogenic (acetate forming) bacteria. These bacteria convert the end-products of hydrolysis into key substrates for methanogenesis (acetate, hydrogen and CO$_2$) and into minor intermediary products (formate, propionate, butyrate, valerate, etc.).

One of key factors in the **anaerobic oxidation** process is the balance between the microorganisms responsible for each step.

**Methanogenesis** The methanogens comprise two physiologically distinct groups of CH$_4$-forming bacteria, namely acetoclastic methanogenic bacteria (converting acetate to CH$_4$) and hydrogen-utilizing methanogenic bacteria (converting hydrogen to CH$_4$). Methanogens are known to be different from the typical bacteria and are classified in a separate kingdom, the Archaea.

CH$_4$ can also be consumed by methanotrophic bacteria under aerobic conditions, converting the CH$_4$ to CO$_2$ either at the air-water or at the air-scum interface where O$_2$ and CH$_4$ are present together (Knowles, 1993) or when CH$_4$ bubbles are released from the sludge.

The CH$_4$ forming microorganisms are strict anaerobes and even small amounts of oxygen can be toxic. The rate of CO$_2$ and CH$_4$ gas production inside the tank is related to temperature and the temperature depends on the water use activities of the household and follows seasonal temperature pattern according to the geographic location (Kinnicutt et al., 1910; Winneberger, 1984). CH$_4$ production can be inhibited by low temperatures...
and insufficient sludge storage capacity (Gray et al., 2011). Sludge and scum accumulation depend on several factors including tank design, user diet, season of the year, and temperature. Winneberger (1984) observed that STs of household with vegetarian occupants developed thin or no scum layers compared to households with a conventional meat-based diet.

**GHG emissions from soil treatment**

However, given that the amount of treatment that occurs in a septic tank is limited and does not generally result in an overall reduction of nutrient loading (Gill et al., 2009), the majority of treatment that occurs in a septic tank treatment system happens in the subsoil of STU beneath the percolation area, see in Section 2.2.4. A continuously fed mature biological clogging zone acts as a potential source of CO₂, CH₄, and N₂O emissions from transformation and degradation of organic and inorganic contaminants in the soil.

Studies have investigated (Magdoff et al., 1974a, b; Gerritse et al., 1995) that aerobic and oxic conditions prevail under trench systems, despite the often saturated nature of the system and the anaerobic character of septic tank effluent. The presence of oxygen, carbon and different forms of N determines the composition of the microbial population (aerobic and anaerobic) responsible for effluent treatment.

**CO₂ and CH₄ emissions from soil treatment units** — soils play an important role in the CH₄ cycle as methanotrophy (oxidation of CH₄) and methanogenesis (production of CH₄) take place, see Figure 2.21. The process of OM decomposition in the soil has been described in detail by Swift et al. (1979). Micro-organisms present in the subsoil use oxygen as their terminal electron acceptor to convert the organic molecules in the percolating water to CO₂, water and energy usually heat. This process of microbial decomposition is dependent on both heat and the availability of oxygen (Kätterer et al., 1998) with oxygen levels usually the limiting factor as these micro-organisms thrive in aerobic conditions. Oxygen supply in the vadose zone occurs by diffusion in the soil atmosphere and therefore the oxygen supply can become limited. Under these conditions anaerobic organisms such as methanogenic bacteria become more dominant. Methanogenic bacteria break down insoluble organic compounds to CO₂ and CH₄ in the absence of oxygen with only limited bacterial growth. Gray (2004) described the process of organic decomposition as occurring in two stages; the first being non-methanogenic in aerobic conditions and the second being methanogenic under anaerobic conditions. The two main processes that occur in the anaerobic stage are hydrolysis and acidogenesis, as described previously.
2.4. GHG EMISSIONS FROM WASTEWATER TREATMENT PROCESSES

Figure 2.21: CO$_2$, CH$_4$ and N$_2$O cycling microbial communities in soil above STU, (Fernandez-Baca et al., 2018).

**N$_2$O emissions from soil treatment** – The source of nitrogen being present in the septic system are mostly human waste, food preparation, hygiene washings and cleaning products. In the ST, most of the nitrogen compounds break down into ammonium (NH$_4^+$) through hydrolysis and ammonium release, see Figure 2.22. The results of this is that the nitrogen content in ST effluent that percolates into the subsoil beneath a percolation area is typically in the form of NH$_4^+$ with some organic nitrogen also present, typically 80 % and 20 %, accordingly (Walker et al., 1973).

During soil treatment, the two main biogeochemical processes to break down further the nitrogen content in the ST effluent are nitrification and denitrification with incomplete denitrification generating N$_2$O emissions.
In the presence of oxygen, the nitrogen content of the ST effluent (NH$_4^+$) typically convert to nitrite (NO$_2^-$) and nitrate (NO$_3^-$) soon after entering the subsoil (Beal et al., 2005) by the processes of nitrification. This aerobic and rapid process occurs in two steps by action of two groups of bacteria, collectively called the nitrifying bacteria. Firstly, Nitrosomonas bacteria transfer the NH$_4^+$ into NO$_2^-$, This reaction immediately followed by the transformation of NO$_2^-$ to NO$_3^-$ by Nictrobacter and Nitrospina bacteria. Nitrifying bacteria are autotrophic genera which reduce CO$_2$, in form of carbonate and bicarbonate, as a source of cellular carbon (Mara and Horan, 2003). Walker et al. (1973) have reported complete or near complete nitrification in the unsaturated zone of the biomat.

In the absence of oxygen, denitrification occurs mainly by ubiquitous facultative heterotrophs which convert NO$_3^-$ to either N$_2$O, nitric oxide gas (NO) or N$_2$. This process is also known as anaerobic or nitrate respiration and carried out by a variety of bacteria such as Alcaligenes, Achromobacter, Micrococcus and Pseudomonas. Not all these bacteria are capable of complete oxidation to nitrogen and their presence is the reason for a variety of gaseous nitrogen being produced. Other environmental parameters such as pH, temperature and NO$_2^-$ and NO$_3^-$ concentrations present regulate the proportion of which nitrogen gases are produced. As well as anaerobic conditions, a source of labile C is required to provide the source of electrons for the oxidation. Optimal conditions for denitrification are generally limited to small anaerobic pockets in the vadose zone or the biomat itself (Wilhelm et al. 1994). Canter and Knox (1985) observed that denitrification is the only process by which concentrations of NO$_3^-$ in the percolating water can be decreased.
2.4. GHG EMISSIONS FROM WASTEWATER TREATMENT PROCESSES

The principal factor in determining the N$_2$O generation potential of wastewater is the amount of N in the wastewater. The variability of N in the influent to the treatment system, as well as the operating conditions of the treatment system itself, will also impact on the generation potential of N$_2$O (USEPA, 2016).

2.4.2 Measured GHG from wastewater treatments

Centralised systems

Wastewater is known to be rich in carbon and nitrogen and wide range of models and measurement techniques have been developed to estimate and measure GHG emissions from large-scale centralised wastewater treatment unit processes. However, on-site DWWTS systems have received less attention compared to centralised wastewater treatment plants (WWTPs) when accounting for GHG releases. Some examples of different methods estimating GHG emissions centralised WWTPs are presented in the following paragraphs and an overview of the emissions can be found in Table 2.2 listing the measurement methods and the systems where the emissions were identified from.

In New Hampshire (US), the CO$_2$, CH$_4$ and N$_2$O emissions from a centralised WWTP were measured using a closed chamber technique from non-aerated surfaces and a bag technique from aerated surfaces, the results of CO$_2$ and CH$_4$ emissions were published by Czepiel et al. (1993) and the N$_2$O emissions in Czepiel et al. (1995). The main sources of emissions for all three gases were the grit tanks, secondary aeration tanks and sludge holding tanks, however; negligible emissions were measured from the primary settling tanks and secondary clarification tanks. The full-scale emissions yield 370 g CO$_2$Eq. m$^{-3}$, 94.9% of the emissions was in the form in CO$_2$, 2.6% in CH$_4$ and 2.5% in N$_2$O.

Johansson et al. (2004) measured CH$_4$ emissions from a constructed wetland in Sweden using the chamber method with multiple floating collars and permanent frames. The spatial and temporal variations were large, ranging between 375 and 1739 mg CH$_4$ m$^{-2}$ d$^{-1}$, but these variations were explained with the recorded environmental factors. The emission values had the strongest correlation with sediment and water temperatures.

Cakir and Stenstrom (2005) compared CO$_2$ and CH$_4$ production from aerobic and anaerobic treatment systems (including sludge digestion) and the losses of dissolved CH$_4$ in digested biosolids and process effluents using a mass balance model. Their main finding showed that above a threshold BOD load (300 mg l$^{-1}$), anaerobic processes will emit less GHGs compared to aerobic processes. The aerobic technology was a conventional activated sludge process and the anaerobic technology was an upflow anaerobic sludge
2. LITERATURE REVIEW

blanket (UASB) reactor. According to the model, for a BOD$_u$ (ultimate carbonaceous biochemical oxygen demand) concentration of 300 mg L$^{-1}$ and 500 mg L$^{-1}$, the total CO$_2$ equivalent gas production in the effluent of the UASB was around 140 mg CO$_{2\text{Eq.}}$ L$^{-1}$ and 108 mg CO$_{2\text{Eq.}}$ L$^{-1}$, while it was 124 mg CO$_{2\text{Eq.}}$ L$^{-1}$ and 220 mg CO$_{2\text{Eq.}}$ L$^{-1}$ at the effluent of the aerobic process. For lower strength influent BOD$_u$ (below 300 mg L$^{-1}$), aerobic processes will emit less GHGs and at higher strengths, anaerobic wastewater treatment is more favourable method.

Sahely et al. (2006) used a life-cycle assessment approach to quantify the total CO$_2$ and CH$_4$ emissions from municipal DWWTSs in Canada for the year 2000. Both on-site emissions (including biological processes used and fossil fuels consumed for energy and heat) and off-site emissions (including production and transmission of fuels and production of electricity) were included in the study. Since this approach was based on assumptions and different values assigned for parameters, to test the sensitivity of the predicted emission of GHGs the study considered potential best and worst case emission scenarios. Estimated on-site CH$_4$ emissions using the IPCC protocol and reported in Canada’s national inventory were approximately 12 times higher than the estimated on-site emission rates in this study. For the year 2000, the on-site emissions from Canadian municipal wastewater treatments were estimated at 1600 Mg CH$_4$ yr$^{-1}$ and 669 100 Mg CO$_2$ yr$^{-1}$, including off-site emissions the estimated total CO$_2$ equivalent emissions rose to 1 048 500 Mg CO$_{2\text{Eq.}}$ yr$^{-1}$.

A tracer dispersion method was applied by Yoshida et al. (2014) to measure plant-integrated and real-time emissions of CH$_4$ and N$_2$O from a WWTP in Denmark during 9 separate field campaigns using two mobile cavity ring-down spectroscopy sampling devices. A wide range of emissions (ranging between 72 and 1865 g CO$_{2\text{Eq.}}$ m$^{-3}$) were detected and the highest emissions were observed during periods experiencing operational problems, such as during foaming events in anaerobic digesters and during sub-optimal operation of biological nitrogen removal in the secondary treatment of wastewater.

To estimate CH$_4$ emissions from a WWTP in France, Kwok et al. (2015) combined a chamber method, which provides insight into individual process emissions, and tracer method, which is suitable for quantifying the total emission of the facility. Direct comparison of total-plant emissions was not possible, because on-site measurements were performed only at process units made by open basins treating wastewater and not applied at the other numerous process units. The estimated CH$_4$ emissions from the full-scale WWTP was 12.5 g CO$_{2\text{Eq.}}$ m$^{-3}$. They found that open basins are not the major source of CH$_4$, but the pretreatment and sludge treatment.

Masuda et al. (2015) in Japan found that the main source of CO$_2$, CH$_4$ and N$_2$O was the consumption of electricity (43.3%), sludge incineration process (8.3%) and the water
treatment process (41.7%), respectively. Three different methods were used to measure emissions from WWTPs: (1) an open chamber method was used to sample gas emitted in the aeration tank, (2) a closed chamber method was used to collect gas emitted on static water surfaces in the equalisation tank, the primary sedimentation tank, the final sedimentation tank, and the disinfection tank, (3) the gas in the ventilation ducts was directly vacuumed and sampled.

In South-West Germany over a period of a year, Alshboul et al. (2016) quantified the amount of CH$_4$ and CO$_2$ discharged with effluent from 9 municipal WWTPs into aquatic ecosystem measuring the CH$_4$ and CO$_2$ concentration upstream and downstream of the effluent and deploying floating chambers and automated bubble traps. They found that effluent significantly altered the physiochemical properties of the stream water and enhanced the CO$_2$ and CH$_4$ emissions downstream of the effluent by a factor of 1.2 and 8.6, respectively. However, the CH$_4$ export to the stream water was only a negligible fraction (0.02%) of the estimated total CH$_4$ emissions during the treatment processes using IPCC protocol.

The IPCC Guidelines excludes CO$_2$ emissions as a result of wastewater treatment from their estimates due to their biogenic origin; hence, only CH$_4$ and N$_2$O are considered as potential sources of emissions from wastewater treatment. For CH$_4$ emissions they established methane correction factors (MCF) for different wastewater treatment and discharge systems. This MCF is multiplied by the BOD or COD loading to calculate the appropriate emission factor. The MCF for septic systems is 0.5. The CH$_4$ emissions calculated over an inventory year [kg CH$_4$ yr$^{-1}$] is the product of the emission factor [kg CH$_4$ kg$^{-1}$ BOD] and organic component [kg BOD yr$^{-1}$] for each treatment-discharge system and for each income group. For N$_2$O, a single emission factor has been established (0.005 kg N$_2$O kg$^{-1}$ N), independent of the wastewater treatment and discharge systems.

Recently the USEPA has provided technical guidance on estimating CH$_4$ and N$_2$O emissions from all domestic wastewater treatment systems in the entire U.S., whereby the total emissions calculated for 2014 were 9.0 MMT CO$_2$Eq. yr$^{-1}$ and 4.8 MMT CO$_2$Eq. yr$^{-1}$, respectively (USEPA, 2016).
Table 2.2: Overview of CO$_2$, CH$_4$ and N$_2$O emissions from centralised WWTSs. In method column, the measurement methods of GHG emissions are listed. Emissions converted to flow based emissions (g gas per m$^3$ wastewater) for comparison. The GHG emissions converted to CO$_2$ equivalent according to their global warming potential (CH$_4$=25, N$_2$O=298).

<table>
<thead>
<tr>
<th>Study</th>
<th>Method</th>
<th>System</th>
<th>g CO$_2$ m$^{-3}$</th>
<th>g CH$_4$ m$^{-3}$</th>
<th>g N$_2$O m$^{-3}$</th>
<th>g CO$_2$Eq. m$^{-3}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Czepiel et al. (1993)</td>
<td>bag technique</td>
<td>aerated grid tank</td>
<td>3.5</td>
<td>5.2 $\times$ 10$^{-2}$</td>
<td>1.5 $\times$ 10$^{-3}$</td>
<td>5.3</td>
</tr>
<tr>
<td>Czepiel et al. (1995)</td>
<td>chamber</td>
<td>non-aerated grit tank</td>
<td>1.0</td>
<td>8.4 $\times$ 10$^{-2}$</td>
<td>1.5 $\times$ 10$^{-4}$</td>
<td>3.1</td>
</tr>
<tr>
<td></td>
<td>chamber</td>
<td>aeration tank</td>
<td>327.3</td>
<td>2.0 $\times$ 10$^{-1}$</td>
<td>2.9 $\times$ 10$^{-2}$</td>
<td>340.9</td>
</tr>
<tr>
<td></td>
<td>sludge sampling</td>
<td>sludge storage tank</td>
<td>19.1</td>
<td>5.7 $\times$ 10$^{-2}$</td>
<td>1.5 $\times$ 10$^{-3}$</td>
<td>21.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>full-scale</td>
<td>350.9</td>
<td>3.9 $\times$ 10$^{-1}$</td>
<td>3.2 $\times$ 10$^{-2}$</td>
<td>370.3</td>
</tr>
<tr>
<td>Johansson et al. (2001)</td>
<td>chamber</td>
<td>constructed wetland</td>
<td>–</td>
<td>-7.3 $-$ 33.8</td>
<td>–</td>
<td>-188 $-$ 845</td>
</tr>
<tr>
<td>Cakir and Stenstrom (2005)</td>
<td>mass balance</td>
<td>aerobic tank</td>
<td>N.A.</td>
<td>N.A.</td>
<td>–</td>
<td>28$^a$, 125$^b$, 508$^c$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>anaerobic tank</td>
<td>N.A.</td>
<td>N.A.</td>
<td>–</td>
<td>45$^a$, 141$^b$, -138$^c$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>full-scale$^e$</td>
<td>N.A.</td>
<td>N.A.</td>
<td>–</td>
<td>2 $-$ 60</td>
</tr>
<tr>
<td></td>
<td></td>
<td>full-scale$^f$</td>
<td>N.A.</td>
<td>N.A.</td>
<td>–</td>
<td>66 $-$ 248</td>
</tr>
<tr>
<td>Daelman et al. (2012)</td>
<td>mass balance</td>
<td>full-scale</td>
<td>–</td>
<td>3.4</td>
<td>–</td>
<td>85</td>
</tr>
<tr>
<td>Yoshida et al. (2013)</td>
<td>tracer dispersion</td>
<td>full-scale</td>
<td>–</td>
<td>1.7 $-$ 31.7</td>
<td>0.1 $-$ 3.6</td>
<td>72 $-$ 1865</td>
</tr>
<tr>
<td>Kwok et al. (2015)</td>
<td>chamber</td>
<td>clarification basin</td>
<td>–</td>
<td>0.003</td>
<td>–</td>
<td>0.08</td>
</tr>
<tr>
<td></td>
<td></td>
<td>aeration basins</td>
<td>–</td>
<td>0.02</td>
<td>–</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>degassing basin</td>
<td>–</td>
<td>0.02</td>
<td>–</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td>tracer dispersion</td>
<td>full-scale</td>
<td>–</td>
<td>0.5</td>
<td>–</td>
<td>12.5</td>
</tr>
<tr>
<td>Masuda et al. (2015)</td>
<td>chamber, life-cycle</td>
<td>full-scale</td>
<td>144.9</td>
<td>1.2</td>
<td>0.4</td>
<td>292.5</td>
</tr>
<tr>
<td>Alshboul et al. (2016)</td>
<td>IPCC</td>
<td>full-scale</td>
<td>–</td>
<td>68.9 $-$ 870.6</td>
<td>–</td>
<td>1723 $-$ 21750</td>
</tr>
<tr>
<td></td>
<td>chamber, bubble trap$^g$</td>
<td>full-scale</td>
<td>0.02 $-$ 0.04</td>
<td>0.003 $-$ 0.13</td>
<td>–</td>
<td>0.1 $-$ 1</td>
</tr>
</tbody>
</table>

$^a$ BOD 100 mg l$^{-1}$  $^b$ BOD 300 mg l$^{-1}$  $^c$ BOD 1000 mg l$^{-1}$  $^d$ on-site biological processes;  $^e$ fossil fuels consumed for energy and heat;  $^f$ production and transmission of fuels and production of electricity;  $^g$ exported dissolved gas with the effluent;
On-site systems

Numerous recent studies on DWWTSs have mainly focused on the attenuation of chemical and biological pollutants and the risk for contamination of groundwater (Gill et al., 2007; Godfrey et al., 2007; Katz et al., 2010; Keegan et al., 2014), wells (Lu et al., 2008; Hynds et al., 2014), or surface waters (Withers et al., 2012; Rosario et al., 2014; Dubber et al., 2016) from septic systems. However, there has been a lack of studies on on-site wastewater treatment systems acting as a source of GHG, the focus of this research.

Estimation of GHG emissions from septic system by USEPA (2016) has been based on a single study conducted by Leverenz et al. (2010) and limited to CH$_4$, by multiplying the U.S. population by the percent of wastewater treated in septic systems and an emission factor 10.7 g CH$_4$ cap$^{-1}$ d$^{-1}$.

Direct techniques to measure gas fluxes from septic tank systems have rarely been reported. Leverenz et al. (2010) and Diaz-Valbuena et al. (2011) using the same data set appear to have been the first to identify the emissions of all three major GHGs (CH$_4$, CO$_2$ and N$_2$O) from eight ST systems and two STUs using a static flux chamber method in California, US. In addition to the flux measurements, the mass flow of gases moving through the household drainage and vent system was determined by measuring the gas flow and concentration at clean-out ports. The septic tank was the primary source of CH$_4$ and CO$_2$ with negligible N$_2$O emissions. Geometric mean values of 11.0 ± 2.2 g CH$_4$ cap$^{-1}$ d$^{-1}$, 33.3 ± 2.7 g CO$_2$ cap$^{-1}$ d$^{-1}$, and 0.005 ± 4.35 g N$_2$O cap$^{-1}$ d$^{-1}$ were reported from the water surface of the ST and 10.7 ± 1.65 g CH$_4$ cap$^{-1}$ d$^{-1}$, 335 ± 2.1 g CO$_2$ cap$^{-1}$ d$^{-1}$, and 0.2 ± 3.6 g N$_2$O cap$^{-1}$ d$^{-1}$ from the venting system, respectively. Emissions from the STUs were deemed to be negligible.

The most recent study by Truhlar et al. (2016) focused on CH$_4$, CO$_2$ and N$_2$O emissions from leach fields and a sand filter at 8 septic systems using a static chamber method in New York, US. Gas concentration and flow in the septic vent system were measured as well. While the majority of GHG emissions escaped through the roof vent as 11 ± 12 g CH$_4$ cap$^{-1}$ d$^{-1}$, 160 ± 3.2 g CO$_2$ cap$^{-1}$ d$^{-1}$, 0.12 ± 3.9 g N$_2$O cap$^{-1}$ d$^{-1}$ (geometric mean ± SD), which were then used as proxy for direct emissions from the ST itself, the STU was found to be a source of N$_2$O emissions of 0.022 ± 1.8 g N$_2$O cap$^{-1}$ d$^{-1}$ (arithmetic mean ± SD) with negligible CO$_2$ emissions. However, an average net-uptake of CH$_4$ of -0.0038 ± 0.0055 g CH$_4$ cap$^{-1}$ d$^{-1}$ was observed within the STU – potentially caused by alterations in the compositions of the prevalent microbial community due to changing water saturation level in the soil (aerobic methanotrophs prevail in drier conditions while
anaerobic methanogens thrive in more saturated conditions). CO₂ fluxes from the sand filter were significantly less than the fluxes over the control field.

Biotic (e.g. mineralization through soil-dwelling animals, bacteria) and abiotic (e.g. soil temperature, water content) drivers control the soil fluxes of GHG emissions. Natural variation of these drivers results spatial and temporal variations of soil fluxes. The existing studies have the limitation of from 3 to 4 months of measurement period (September to December 2009, Diaz-Valbuena et al. (2011); June to August 2014, Truhlar et al. (2016)) and only recording a limited number of measurements per site (between 2–5), which did not take into account any expected seasonal variation of the emission rates. Equally, the measurements would not have captured any potential diurnal variation. Additionally, these studies neglected the spatial variation of emissions from the STU.

2.4.3 Relative contribution to global anthropogenic budget

IPCC (2013) estimated that the post-consumer waste and wastewater contribution to the total global anthropogenic emissions in 2010 was less than 5% (see Figure 2.23). The total global emissions from waste and wastewater almost doubled between 1970 and 2010. Waste and wastewater accounted for 1.5 Gt CO₂Eq. GHG emissions, 90% of these emissions are accounted for CH₄ from municipal solid waste disposal and from wastewater. The main source of waste GHG emissions is solid waste disposal on land, the second largest source is wastewater handling and minor emissions of CO₂ from incineration of waste containing fossil carbon (Bogner et al., 2007). IPCC (2013) considers wastewater N₂O as minor source and excludes wastewater CO₂ emissions due to their biogenic origin.

Waste generation including wastewater is closely linked to the increasing population, prosperity and urbanisation. However, these global estimates have a high uncertainty and this report highlights the importance of the more direct measurements of GHG emissions from the waste sector including wastewater management to validate the models used for the estimates.
2.5. REVIEW OF METHODOLOGY

2.5.1 Overview of GHG detection methods

There are a number of commonly used methods to assess GHG emissions from soil–plant systems and water bodies depending on the pathway of interest and the area of the study site. From soil–plant systems, flux chambers or eddy–covariance towers are the most common methods to measure GHG emissions. The eddy–covariance method integrates fluxes over a footprint area of up to several hundred square metres and is not applicable for measuring localised GHG emissions from on-site wastewater treatment units. Flux chambers covering a surface are in the range of square centimetres to square metres have been deployed successively over terrestrial and aquatic ecosystems. For diffusive emission pathways from aquatic surfaces fluxes can be modelled using thin boundary methods or directly obtained from chamber measurements using manual or automatic floating chambers (St. Louis et al., 2000). For ebullition pathways, rates can be directly measured using floating chambers, acoustic surveys or funnel traps (DelSontro et al., 2010; Bastviken et al., 2011; Maeck et al., 2013). Thin boundary layer models cannot be used to quantify
the ebullition pathway and acoustic surveys or funnel traps cannot be used effectively in shallow water bodies and both methods can not be adapted for soil-plant systems (Lorke et al., 2015). The flux chamber method is, therefore, the most commonly used method for measuring GHG emissions from soil-plant systems, as well as directly from water surfaces over a small footprint. For this study an automated soil gas flux system was tailored to measure fluxes from the water surface in the ST and as well as from the soil over the STU. Gas analysers were also used to measure the dissolved CO₂ and CH₄ concentrations in the wastewater as well as the gases escaping from the vent system outlets.

Chemical methods

There are only a few purely wet chemical methods for the analysis of GHGs in common use. Their disadvantage is that they have much lower sensitivities compared to optical and chromatographic methods and usually they are labour intense. However, they are relatively simple and inexpensive. For example, CO₂ can be trapped on basic absorbent or in basic solution – chemical absorption and gravimetry by weighing the absorbent before and after exposure for a given amount of time.

Optical methods

Optical methods of analysis exploit the interaction of radiant energy with matter. Measurement techniques are based upon the radiation absorption for the quantification of GHGs because those gases absorb radiation at very specific and unique wavelengths. The amount of energy in a beam of radiation of a particular wavelength that passes through a sample matrix that contains an absorbing gas is quantitatively reduced in a fashion that depends directly upon the path length and the concentration of the absorbent. Visible and ultraviolet methods are usually considered separately from those methods that use the absorption of radiative energy at the infrared wavelengths (Crill et al., 1993; Oertel et al., 2016).

Non-Dispersive Infrared (NDIR) techniques use a gas filter to selectively absorb IR radiation passing through a gas sample. An NDIR analyser relies on the same principle of IR absorption, that makes GHGs an important issue with regards to global warming in the first place. An IR analyser consists of an IR source at one end and an IR detector, separated by a gas cell. The gas of interest is passed through this cell and absorbs some of the infrared radiation coming from the source. The detector converts the amount of IR reaching it to a usable signal, such as a voltage. Hence the concentration of CO₂ changes in
the sample, the signal from the detector changes. By flowing a gas with a known amount of CO$_2$ through the cell, the analyser is calibrated such that the voltage output from the detector can be converted into amounts of CO$_2$.

**Off-Axis Integrated-Cavity Output Spectroscopy (Off-Axis ICOS)** utilizes a high-finesse optical cavity such as an absorption cell. Unlike conventional multi-pass arrangements, which are typically limited to path lengths less than two-hundred meters, an Off-Axis ICOS absorption cell effectively traps the laser photon so that, on average, they make thousands of passes before leaving the cell. As a result, the effective optical path length may be several thousands of meters using high-reflectivity mirrors and thus the measured absorption of light after it passes through the optical cavity is significantly enhanced.

**Fourier Transform Infrared Spectroscopy (FTIR)** In an FTIR spectrometer, incoming infrared light is split into the two paths of a two-beam interferometer in which one path contains a moving optical element, typically a mirror. When the beams are combined at an infrared detector, constructive and destructive interference produces a modulated signal which is a function of the optical path difference between the two beams. The digitalised record of this signal, the interferogram, is converted into a spectrum by a complex Fourier transform. In contrast to a prism or grating spectrometer which operates by selecting only a small fraction of the light entering its input slit, the light entering an FTIR spectrometer falls on the detector at all times. The other components of the systems are an infrared source, an optical system which defines the sampling region between source and spectrometer, and a computer which not only perform the data acquisition and spectrum generation, but can be programmed to aid in the analysis of spectra as well.

**Chromatographic methods**

Gas chromatography is a separation method by exploiting selective adsorption / desorption characteristics of compounds in which gas mixtures flow over a material that retains some components more than others, so different components flow over the material at different speeds. The gas is separated into individual chemical components which can then pass through a detector, which varies in output for each component in the mixture. This is called a chromatogram. By measuring the height of each peak in a chromatogram, the amount of a component can be calculated. Chromatography is the preferred method for CO$_2$, CH$_4$ and N$_2$O since it is well suited for small samples with small amounts of the gas of interest.
2.5.2 Chamber method

Chamber techniques, also called enclosure-based measurements earlier (Livingston and Hutchinson, 1995), are widely used in terrestrial (Kutzbach et al., 2007; Forbrich et al., 2010) and aquatic studies (Bastviken et al., 2011; Lorke et al., 2015) of GHG emissions. They are relatively low in cost, simple to operate, resulting discrete observations in space and time. Different shapes are used either rectangular or cylinder for the chamber where the emitted gases can accumulate. The change of mixing ratio can be analysed with various gas sensors, e.g., gas chromatography, IR-spectrometry including NDIR and FID with and without pumps, Cavity-Ring-down spectrometry or photo-acoustics.

2.5.3 Eddy covariance

Eddy covariance is a direct micro-meteorological method using vertical turbulences to analyse the turbulent heat and gas exchange between the terrestrial surface (can include aquatic systems) and atmosphere (Launiainen et al., 2005). A 3D ultrasonic anemometer and a gas analyser attached to a tower at least a 2 m height. With the eddy covariance method, the whole surface area is being integrated (i.e. a several hundreds of metres) completely covering biosphere and atmosphere to determine net ecosystem exchange and therefore this approach is not currently suitable for analysing GHG emissions from individual STUs.

2.5.4 Remote sensing methods

Remote sensing from satellites may deliver information on GHG soil emissions. One approach is to estimate tropospherical, near-surface CO₂ and CH₄ concentrations based on measurements of the intensity of the reflected sunlight in small wavelength bands up the visible and short-wavelength IP portion of the spectrum in order to gain high spatial resolution. However, the spatial resolution is still at several square kilometres (Oertel et al., 2016) which would not be able to pick up individual DWWTSs.
3

Construction and Instrumentation of Field Sites

This chapter describes the selected field sites that were constructed, instrumented and sampled. Section 3.1 introduces the field sites and their wastewater treatment systems. Section 3.2 details the different GHG measurement setups. Section 3.3 describes the meteorological monitoring setup.

3.1 Field sites

In total, 4 sites were intensively monitored over the period of this research (February 2015 to August 2018) to have a better understanding of and quantity of GHG emissions from on-site wastewater treatment systems. All the 4 sites are located in the Republic of Ireland (see Figure 3.1) and were domestic houses in rural locations, owned by the residents who carried on with their day-to-day activities as usual during the monitoring period. The sites were chosen according to different criteria as part of previous/ongoing research projects. One of the 4 sites, Kilbeggan (KB), was constructed over 20 years ago and had previously been used as a monitoring sire for a septic tank soakaway in an EPA project (2010-W-LS-3) entitled “Assessment of disposal options for treated wastewater from single houses in low permeability subsoils” (Gill et al., 2015). The other 3 sites (Kilmallock KM, Crecora CC, Louth LO) were constructed and instrumented by the Department of Civil, Structural and Environmental Engineering at Trinity College Dublin between 2015 and 2016. KM and CC sites were set-up in the frame of the doctoral researcher project - “Modelling of soil biomass for on-site wastewater treatment” - which aims to determine the impact of the biomat that forms at the infiltrative layer at the base of percolation trenches on pollutant
transport and attenuation. LO was designed as part of project on “Evapotranspiration willow systems for on-site wastewater treatment in areas of low permeability soils”. Several sites were investigated during the course of these projects to end up with the final three households chosen with desired characteristics with details of the selected sites and systems given in Table 3.1.

![Figure 3.1: Locations of the field sites; Kilbeggan (KB), Kilmallock (KM), Crecora (CC) and Louth (LO).](image)

**The climate** is classified as maritime CFb (warm temperate, fully humid, warm summer) after Köppen-Geiger ([Kottek et al., 2006](#)) at all four sites. The mean annual temperature is 10°C with mean seasonal minimum and maximum between 4°C and 15°C in winter and summer, respectively; average annual rainfall ranges from 700 mm in Louth up to 1200 mm in the south western sites without significant seasonal variation and 1250 h total annual sunshine ([Walsh, 2012](#)).
Table 3.1: Characteristics of the selected study sites and wastewater treatment systems.

<table>
<thead>
<tr>
<th>Study sites</th>
<th>KB</th>
<th>KM</th>
<th>CC</th>
<th>LO</th>
</tr>
</thead>
<tbody>
<tr>
<td>County</td>
<td>Westmeath</td>
<td>Limerick</td>
<td>Limerick</td>
<td>Louth</td>
</tr>
<tr>
<td>Aquifer Category</td>
<td>Rkd&lt;sup&gt;a&lt;/sup&gt;</td>
<td>Li&lt;sup&gt;b&lt;/sup&gt;</td>
<td>Rkd&lt;sup&gt;a&lt;/sup&gt;</td>
<td>Pu&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td>Groundwater vulnerability</td>
<td>H&lt;sup&gt;d&lt;/sup&gt;</td>
<td>M&lt;sup&gt;e&lt;/sup&gt;</td>
<td>H&lt;sup&gt;d&lt;/sup&gt;</td>
<td>L&lt;sup&gt;f&lt;/sup&gt;</td>
</tr>
<tr>
<td>Topsoil (above 25 cm)</td>
<td>BminDW&lt;sup&gt;g&lt;/sup&gt;</td>
<td>BminDW&lt;sup&gt;g&lt;/sup&gt;</td>
<td>BminDW&lt;sup&gt;g&lt;/sup&gt;</td>
<td>AminPD&lt;sup&gt;h&lt;/sup&gt;</td>
</tr>
<tr>
<td>Subsoil (below 25 cm)</td>
<td>TLI&lt;sup&gt;i&lt;/sup&gt;</td>
<td>ClLo&lt;sup&gt;j&lt;/sup&gt;</td>
<td>SaLo&lt;sup&gt;k&lt;/sup&gt;</td>
<td>IrSTLPS&lt;sup&gt;l&lt;/sup&gt;</td>
</tr>
<tr>
<td>Land use of surrounding area</td>
<td>Pasture land</td>
<td>Garden</td>
<td>Pasture land</td>
<td>Garden</td>
</tr>
<tr>
<td>No. of occupants</td>
<td>2-3</td>
<td>4</td>
<td>4</td>
<td>4</td>
</tr>
<tr>
<td>Primary treatment</td>
<td>Single compartment ST</td>
<td>Two-compartment ST</td>
<td>Two-compartment ST</td>
<td>Two-compartment ST</td>
</tr>
<tr>
<td>Capacity (m&lt;sup&gt;3&lt;/sup&gt;)</td>
<td>2.6</td>
<td>4.8</td>
<td>4.8</td>
<td>3.1</td>
</tr>
<tr>
<td>Desludged</td>
<td>2012</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>HRT&lt;sup&gt;n&lt;/sup&gt;(d)</td>
<td>7.2</td>
<td>10.3</td>
<td>10.3</td>
<td>10.7</td>
</tr>
<tr>
<td>Secondary treatment</td>
<td>Coconut filter</td>
<td>RBC</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>Effluent</td>
<td>Gravity flow</td>
<td>Pumped</td>
<td>Gravity flow</td>
<td>Gravity flow</td>
</tr>
<tr>
<td>Soil treatment unit</td>
<td>Soakaway</td>
<td>2-trench - PE&lt;sup&gt;m&lt;/sup&gt;</td>
<td>2-trench - PE&lt;sup&gt;m&lt;/sup&gt;</td>
<td>Willow</td>
</tr>
<tr>
<td>Area (m&lt;sup&gt;2&lt;/sup&gt;)</td>
<td>6</td>
<td>180</td>
<td>180</td>
<td>2 x 247</td>
</tr>
<tr>
<td>K&lt;sub&gt;f&lt;/sub&gt;&lt;sup&gt;r&lt;/sup&gt;(m d&lt;sup&gt;-1&lt;/sup&gt;)</td>
<td>1.39</td>
<td>0.32</td>
<td>0.18</td>
<td>0.08</td>
</tr>
</tbody>
</table>

<sup>a</sup> Regionally Important Aquifer - Karstified Bedrock (diffuse); <sup>b</sup> Locally Important Aquifer - Bedrock which is Moderately Productive only in Local Zones; <sup>c</sup> Poor Aquifer - Bedrock which is Generally Unproductive; <sup>d</sup> High; <sup>e</sup> Moderate; <sup>f</sup> Low; <sup>g</sup> Deep well drained mineral soil derived from mainly basic; <sup>h</sup> Mineral poorly drained - Mainly acidic; <sup>i</sup> Limestone till; <sup>j</sup> Clay loam; <sup>k</sup> Sandy loam; <sup>l</sup> Irish Sea Till derived from Lower Palaeozoic sandstones and shales; <sup>m</sup> Hydraulic Retention Time; <sup>n</sup> Primary effluent; <sup,o</sup> Secondary effluent; <sup>p</sup> Field Saturated Conductivity.
3.1.1 Kilbeggan

The KB research site is located in County Westmeath, see Figure 3.2. The site is homogeneously vegetated grassland previously used for cattle grazing lying in an area of high groundwater vulnerability and is characterized by high permeability, gravelly SAND subsoil according to British Standard classification BS 5930 (BSI, 1999) consisting of till derived mainly from limestone. The DWWTS, receiving effluent from a single household with a fluctuating number of occupants averaging 2 to 3, was constructed more than 20 years ago. The system consists of a single compartment septic tank and a subsequent soakaway, see Figure 3.3 for site layout.

![Figure 3.2: Kilbeggan site.](image)

**Primary treatment** is single-compartment, concrete ST fed by a single gravity flow effluent pipe from the household and has a total capacity of 2.6 m$^3$ with an estimated mean hydraulic retention time of 7 d. The septic tank was last desludged 3 yr before the start of this study, see Figure 3.4.

**STU** is a soakaway pit back-filled with stone or rubble for effluent disposal, see Figure 3.4. This soakaway distributes an average 360 L d$^{-1}$ of effluent over a total area of approximately 6 m$^2$ from a single effluent pipe. The soakaway had previously been the subject of an intensive study which aimed to delineate the effluent plume and determine pollutant attenuation ([Keegan et al., 2014](#); [Gill et al., 2015](#)). It had been instrumented with soil moisture samplers (suction cup lysimeters) throughout its depth. The effluent
from the ST was found to move rapidly through the subsoil in a predominantly vertical direction following the overall gradient of the site. On-site falling-head percolation tests yielded a field saturated conductivity $K_{fs}$ of 1.39 m d$^{-1}$. There was little evidence of the effluent plume extending laterally (apart from along the topographic gradient), consistent with the free draining subsoil characteristics. Despite the high permeability of the site, most of the organic and phosphorus concentrations in the effluent were reduced within the biologically active zone of the soakaway which develops at the infiltrative surface over time. Equally, the unsaturated conditions in the soil gave rise to high levels of nitrification which occurred rapidly within the receiving subsoil, with 98% of the inorganic-N present as NO$_3$–N indicating a potential source of N$_2$O emissions (Gill et al., 2009, 2013).
3.1.2 Kilmallock

The KM research site is located in County Limerick, see Figure 3.2. The site is homogeneously vegetated grassland at the back-garden lying in an area of medium groundwater vulnerability and is characterized as clay loam according to British Standard classification BS 5930 (BSI, 1999) consisting of till derived mainly from limestone. The DWWTS, receiving effluent from a single household with 4 occupants, was constructed in September 2015. The system consists of a two-chamber septic tank, pump sump with two pumps each feeding separate main rising discharge pipe. One pipe is connected to a baffled attenuation tank and which feeds two trenches with primary effluent from the ST and the other pipe discharges to a secondary treatment system with coco husk filtering media that feeds another two trenches with secondary effluent, see Figure 3.5 and 3.6.
3.1. FIELD SITES

Figure 3.5: Soil treatment unit at KM site.

Figure 3.6: Layout of the Kilmallock site and treatment system.
Primary Treatment was a two-chamber prefabricated concrete ST (Aswasep Septic Tank NS 4 S Tank, Molloy Precast Products Ltd., the Republic of Ireland) that was designed and constructed within the guidelines set out in the EPA Code of Practice (EPA, 2009) and it was tested to EN 12666-1 at PIA in Aachen, Germany with the capacity of 4760 L. From the second compartment of the ST, water passes by gravity to a pump sump with two pumps feeding the rising mains. The pumps (DOC3 Lowara, Xylem Water Solution Ltd., the Republic of Ireland, 145 L min\(^{-1}\), head < 7 m, suspended solids up to 10 mm, 220 V 50 Hz 2 poles, 2850 rpm) were controlled by a float switch which triggered one pump at the time once a certain level of effluent was reached in the chamber, see Figure 3.7. The two-pump system was used in order to equally distribute the wastewater between two pairs of trenches, as described previously.

![Figure 3.7: At Kilmallock, two-compartment ST and baffled tank with two pumps.](image)

Secondary Treatment was provided by a media filter (Ecoflo, Premier Tech Aqua, Premier Tech) that uses shredded coconut husk fibre as filter material (size range of mm to cm) acting as a physical barrier to retain remaining suspended solids and providing a high specific surface area for promoting microbial growth to biologically treat the passing effluent, see Figure 3.8. As primary effluent is dosed intermittently into the media filter and distributed evenly across the entire filter surface area via a tipping bucket mechanism, both aerobic and anaerobic degradation processes are facilitated within the unit, with the filter medium itself being a potential source of carbon, potentially enabling denitrification of previously nitrified effluent. Being a by-product derived from the coconut industry, coconut husk as a natural organic filtering media is generally more sustainable and environmentally friendly than other material traditionally used in on-site secondary filtration systems, such as sand, textile or foam media. After its expected life time of approximately 15 years, the coconut husk is be replaced by service provider and digested to biogas or composted off-site.
3.1. FIELD SITES

STU consisted of four perforated sewage pipes (MFP HITEC length 18 m, diameter 110 mm) that were laid 2.5 m apart in gravel in pre-excavated channels at a nominal depth of 0.5 m, see Figure 3.9. The base of the trenches were filled with 250 mm of washed gravel (20–30 mm diameter) on which the pipes were then laid at a gradient of 1 in 200. The pipes were then covered with a 100 mm layer of washed gravel. On the gravel, geotextile (Terram 1000) was placed and backfilled with the excavated soil. The total area of the STU was 180 m (18 m length x 10 m width). The STU was designed and constructed within the guidelines set out in the EPA Code of Practice (EPA, 2009).

Figure 3.9: At Kilmallock, STU construction.
3.1.3 Crecora

The CC research site is located in County Limerick. The site is homogeneously vegetated pasture land previously used for horse grazing lying in an area of high groundwater vulnerability and is characterized by high permeability, gravelly SAND subsoil according to British Standard classification (BSI, 1999) consisting of glacial till derived mainly from limestone, Figure 3.2. The DWWTS, receiving effluent from a single detached house with 4 occupants, was constructed in April 2016. The system consists of a two-chamber septic tank, with two outlet pipes; one feeding two trenches with primary effluent from the ST and the other discharging to an RBC secondary treatment system that discharges secondary treated effluent into another two trenches, see Figure 3.10. In order to avoid the seasonal flooding observed with the previous system (one chamber ST with a soakaway), all rainwater, surface water, and runoff associated drainage were diverted by locating the new tank several meters away from a large concreted area behind the main house with sufficient natural soil area for drainage infiltration in between.

![Image](image_url)

Figure 3.10: Layout of the Crecora site and treatment system.

**Primary Treatment** was a two-chamber prefabricated concrete ST (Aswasep Septic Tank NS 4 S Tank, Molloy Precast Products Ltd., the Republic of Ireland) that was designed and constructed within the guidelines set out in the EPA Code of Practice (EPA, 2009) and it was tested to EN 12666-1 at PIA in Aachen, Germany with the capacity of...
3.1. FIELD SITES

Figure 3.11: (a) installation of ST, (b) RBC, (c) STU and (d) the completed STU at CC.

4760 L, see Figure 3.11a. From the second compartment of the ST, the effluent was equally split by a tipping bucket distribution box (Molloy Precast Products Ltd., the Republic of Ireland). One half was directly fed by gravity to another tipping bucket which distributed the effluent evenly between two trenches and the other half was fed into a secondary treatment system.

**Secondary Treatment** was a rotating biological reactor (RBC, Klargester BioDisc, Kingspan Ltd., UK), see Figure 3.11a. The RBC was made of glass-fibre reinforced polyester and consists of an integrated primary settling chamber, a two-stage biozone, and a secondary clarification chamber, see Figure 3.12. In the primary settling chamber,
remaining suspended solids from the septic tank effluent can settle down at the bottom and
the liquid fraction passed through a submerged baffle into the first stage of the biozone.
In the biozone, corrugated polypropylene discs with an overall surface area of 72.5 m² were
mounted as medium in two separate stages on a horizontal shaft (IAB, 2001). The shaft
rotates slowly (approximately 2 rpm) and effluent is transported from the first to the sec-
ond stage by means of a cup connected to the shaft. This design limits the maximum flow
rate through the RBC to about 50 l h⁻¹ and, thus, hydraulically buffers peak flows into
the STU. The secondary clarification chamber allows for final sedimentation of remaining
suspended solids and excess biofilm flocs detached from the media due to shear forces.

Figure 3.12: (a) schematic drawing of Klagester BioDisc RBC and (b) biofilm growth on
the media after 24 month of use.

**STU** consisted of four perforated sewage pipes (MFP HITEC length 18 m, diameter
110 mm) that were laid 2.5 m apart in gravel in pre-excavated channels at a nominal depth
of 0.5 m, see Figure 3.11c. The base of the trenches were filled with 250 mm of washed
gravel (20–30 mm diameter) on which the pipes were then laid at a gradient of 1 in 200. The
pipes were then covered with a 100 mm layer of washed gravel. On the gravel, geotextile
(Terram 1000) was placed and backfilled with the excavated soil. The total area of the
STU was 180 m² (18 m length x 10 m width), see Figure 3.11d. The STU was designed and
constructed within the guidelines set out in the EPA Code of Practice (EPA, 2009).
3.1. FIELD SITES

3.1.4 Louth Site

The site is located in County Louth in North-East of the Republic of Ireland, see Figure 3.11. The top and subsoil are classified as AminPD (mineral poorly drained - mainly acid) and IrSTLPSS (Irish Sea till derived from lower palaeozoic sandstones and shales), respectively. The underlying aquifer is a poor aquifer, bedrock which is generally unproductive and the groundwater vulnerability is low.

The site was designed and constructed as part of project on “Evapotranspiration willow systems for on-site wastewater treatment in areas of low permeability soils” based on extensive field trials on 13 full-scale sealed willow systems in County Wexford, Leitrim and Limerick by the Environmental Engineering Research Group in Trinity College Dublin Curneen and Gill (2015, 2016).

The wastewater treatment system was built in 2016 and used by a single household with 4 full-time inhabitants (i.e. 4 PE) and it is located the back garden of the family house. The design was based upon 100 l cap$^{-1}$ d$^{-1}$ (litres per person per day) wastewater production, which is considerably lower than the figure of 150 l cap$^{-1}$ d$^{-1}$ assumed in the Code of Practice (EPA, 2009). On this basis, the house was fitted with water saving devices, such as dual flush toilets, low flow shower heads and tap aerators to achieve the lower wastewater production.

The system consists of a two-chamber polyethylene septic tank and two sealed willow evapotranspiration basins, see Figure 3.13 and 3.14. The wastewater effluent from the ST is discharged to the willow system by gravity where it is distributed via a pipe network situated in a gravel layer at the bottom of the basin. The design is based upon wastewater production figures of 100 litres per person per day which is a realistic figure for a single household fitted with water saving devices. The system incorporated an inspection well, in which the water level is monitored and allows for pumping out of the system, as well as an overflow pipe. A bund is constructed along the perimeter of the willow system surface to create extra storage in case of surface flooding.

**Primary Treatment** was a ST (Premier Tech Aqua Ltd.) that was designed and constructed within the guidelines set out in the EPA Code of Practice (EPA, 2009) and it was tested to EN 12666-1 at PIA in Aachen, Germany. The ST was made of polyethylene with 2 chambers and capacity of 3.6 m$^3$ and hydraulic retention time of 10.7 d.

The effluent from the septic tank was gravity fed to a distribution box, where the effluent is equally divided between ET treatment systems in parallel.
It should be noted that trials carried out by Curneen and Gill (2014) have demonstrated no advantage of discharging secondary effluent onto the willows compared to primary treated effluent; in fact willows dosed with primary treated effluent showed a slight enhancement on evapotranspiration rates (mean 7.21 mm d$^{-1}$) compared to the willow trees receiving secondary treatment effluent (mean 5.61 mm d$^{-1}$).

The size of the evapotranspiration system was designed for 4 PE, the minimum recommended size of 500 m$^2$, the area is divided into 2 basins, see Figure 3.15a and 3.15b for top view and cross section, respectively. The basins of the willow system was excavated to a minimum depth at any point of 1.8 m with 8.5 m width and 29 m length. The basin was lined an impermeable membrane (0.5 mm thickness butyl rubber), which is surrounded on both sides by a geosynthetic barrier with properties as specified in Table 3.2.

<table>
<thead>
<tr>
<th>Property</th>
<th>Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>Impermeable Membrane Thickness</td>
<td>0.5 mm</td>
</tr>
<tr>
<td>Strength at break</td>
<td>8 MPA</td>
</tr>
<tr>
<td>Elongation at break</td>
<td>350 %</td>
</tr>
<tr>
<td>Tear Resistance</td>
<td>30 N per m</td>
</tr>
<tr>
<td>Puncture Resistance</td>
<td>120 N</td>
</tr>
<tr>
<td>Geosynthetic Barrier Thickness</td>
<td>1.5 mm</td>
</tr>
<tr>
<td>Mass per unit area</td>
<td>180 g per m$^2$</td>
</tr>
</tbody>
</table>
3.1. FIELD SITES

The 300 mm gravel layer consisting of 20 mm washed gravel was placed on top of the geosynthetic barrier at the base of the willow system. In each basin, two rows of percolation pipes (110 mm diameter) were laid in the middle of the gravel layer at a maximum distance of 4 m apart to distribute the wastewater. The piping was covered with another layer of geosynthetic membrane to prevent the ingress of soil particles down into the distribution layer.

An inspection well was installed to allow for pumping out (if necessary) as well as continuous monitoring of the water level within the system. The inspection well comprises of a 300 mm diameter perforated plastic pipe wrapped in geosynthetic layer, which is placed on top of concrete tiles at the base of the system to protect the impermeable membrane and to prevent the ingress of soil prior to backfilling. The well is located approximately 3 m from the opposite end to where effluent is pumped in. Once the gravel layer, wastewater distribution network and inspection well were established the original soil was backfilled into the basin. A 0.3 m bund was constructed around the perimeter at the surface of the willow system with impermeable membrane and geosynthetic liner as overflow protection. Finally, a weed-proof permeable membrane was used to cover the area of each system.
Planting of willow cuttings took place in March 2016, 3 months after construction, at a density of $3 \text{ m}^{-2}$, rows were planted 0.75 m apart with the cuttings spaced at 0.45 m in each row. Three different willow varieties were used; Tora, Tordis and Olof - all cultivars of *Salix viminalis*. Cuttings were 20 - 30 cm in length with a minimum diameter of 9 mm in order to maximise the chance of establishment.
Additional instrumentation: The wastewater flow into each willow system and the water level in each basin were monitored continuously. For measuring the water flow, tipping bucket flow recorders were used and in a stilling well, the water level was measured with using an OTT Orpheus Mini groundwater datalogger (accuracy ± 0.05%). Additionally, the flow out of the overflow was monitored using a tipping bucket flow recorder.

3.2 GHG measurement setups

The GHG measurements were conducted from February 2015 to August 2018 at the above mentioned field sites using different set-ups, see Figure 3.16. Data from KM site late 2015 and early 2016 was excluded from the final study due to the extreme flooded conditions during that winter. During autumn and winter 2016, there were ongoing complications with the soil flux chambers and this dataset was also excluded from the final analysis. One of the most common methods for measuring the GHG emissions from soil-plant systems and as well as from water surface is to use flux chambers (see Section 2.5.2). The GHG fluxes were directly sampled measured from the water surface of STs as well as from the STUs using an automated flux chamber system (LI-8100 Automated Soil CO₂ Flux System, LI-COR) with an additional gas analyser (UGGA Ultraportable Greenhouse Gas Analyser, model 915-0011, manufactured by Los Gatos Research) for CH₄, see Figure 3.17. Measurements of dissolved gas concentration in the STs and gas-phase concentrations within the vent systems were also directly sampled. Additional gas samples were collected and transported back to the laboratory for further analyses of CH₄ and N₂O.
3. SITES AND INSTRUMENTATION

Figure 3.17: A - survey chamber (LI-8100-103), B - long-term opaque chamber, C - long-term transparent chamber, D - integrated flux analyser system with an long-term opaque and clear chambers, multiplexer, analyser and UGGA - source: https://www.licor.com

3.2.1 GHG measurements from septic tank

Two different setups were used to measure CO$_2$ and CH$_4$ gas fluxes from the ST; (i) discrete survey measurements carried out manually whilst on site and (ii) diurnal measurements carried out by automated survey flux chamber. Additional to the flux measurements, water samples were collected to estimate the dissolved CO$_2$ and CH$_4$ concentrations.

Discrete survey measurements

In order to measure GHG fluxes from the water surface of ST, a sampling setup was adopted from (Leverenz et al., 2010), where a collar was placed into the septic tank to hold the chamber and create a gas loop between the water surface and gas analyser during measurements. This collar was composed of a rigid PVC pipe (inner diameter 20.3 cm, outer diameter 21.3 cm O.D. and length 19 cm) and supported by three legs (length 210 cm) going to the bottom of the tank, see Figure 3.18a. The collars were placed such that they extended from beneath to above the water level of the ST, see Figure 3.18b. These inserts were left in place for the duration of the experiment to prevent disturbance in the sludge of the septic tank.

A survey chamber (LI-8100-103 20 cm Survey Chamber, LI-COR Biosciences, Inc.), that was connected to an analyser control unit with two sets of LI-COR extensions (length:
3.2. GHG MEASUREMENT SETUPS

Figure 3.18: (a) stand with the collar that was inserted into the ST and (b) deployment of the survey chamber over the ST.

15 m), was lowered down to the collar in the ST. The lids of the ST was opened in each compartment. One set of the extensions connected the gas lines, the other set of extensions was controlling the survey chamber and providing the gas channel for lowering down and lifting up the survey chamber over the collar between measurements. The survey chamber manually was placed to the collar prior to the measurement and a pressure/vacuum air flow system expands and contracts a bellows to raise and lower the chambers over the collar to conduct the flux measurement. CO$_2$ fluxes were measured by the non-dispersive infrared CO$_2$ gas analyser unit of an automated soil gas flux system (LI-8100 Automated Soil CO$_2$ Flux System, LI-COR), see technical details in Appendix A. In order to measure CH$_4$ fluxes as well, this CO$_2$ flux system was extended with an additional gas analyser (UGGA Ultraportable Greenhouse Gas Analyser, model 915-0011, manufactured by Los Gatos Research). On the occasions when Los Gatos analyser was available, its gas inlet port was connected with the LI-COR analyser unit outlet port and the gas outlet from the Los Gatos was connected to gas port of the extension going to the survey chamber. During these flux incubations, that were at least 3 minutes of duration, changes in CO$_2$ and CH$_4$ concentrations were observed - increase in concentrations indicating emission and used to calculate fluxes.
At KM and CC, CO₂ fluxes were measured on 13 occasions and CH₄ fluxes on 8 occasions between May 2017 and August 2018. No flux measurements from the ST were conducted at KB and LO.

**Diurnal survey GHG flux measurements**

Additionally to the discrete survey GHG flux measurements, CO₂ and CH₄ fluxes were recorded at two different sampling dates with 5-minute frequency in order to have a better understanding of the diurnal variation of the fluxes. The same system was used as described above. At CC site the diurnal measurements were set up in the 2nd chamber between 2017-11-20 16:30 and 2017-11-21 12:00. At KM site the diurnal measurements were set up in the 2nd chamber between 2018-07-24 19:00 and 2018-07-25 19:00.

**Dissolved GHG concentrations** were measured using the headspace method (Hope et al., 1993) and UGGA (Ultra-Portable GGA, model 915-0011, Los Gatos Research). 100 ml water samples were collected in a 250 ml glass bottle, the bottle was sealed with the cup and attached and inlet and outlet pipe. The pipes were directly connected to the UGGA, the analyser bubbled out the dissolved gases to enable the equilibrium concentration of the gases in the bottle system to be measured, see Figure 3.19.

![Figure 3.19: Bubbling water samples to measure dissolved gas concentration of CH₄ and CO₂. The arrows indicate the gas flow direction.](image)

Prior to the GHG flux and dissolved GHG concentration measurements, dissolved oxygen (DO), pH, electrical conductivity (EC) and water temperature (T) were measured at 15
cm in both chambers of the ST using a WTW multi-parameter portable meter (WTW ProfiLine Multi 3320, WTW GmbH, Germany). For the technical specifications of the probes see Appendix B.1.

All pH and EC readings were automatically temperature compensated by the hand-held meter.

### 3.2.2 GHG flux over soil treatment unit

Two different methods were used to measure CO$_2$ and CH$_4$ soil fluxes from the STU; (i) discrete survey measurements carried out manually whilst on site and (ii) diurnal measurements carried out by automated long-term flux chambers. Additional to the flux measurements, gas samples were collected and carried to the laboratory to measure CH$_4$ and N$_2$O flux with a gas chromatography.

The same automated gas flux system was used to measure the fluxes from STU. The collars were installed in the ground (approximately 0.05 m), leaving approximately 0.1 m above ground on which flux chambers were placed creating a seal between the collar and the chamber. For control measurements additional collars were installed in the region surrounding the STU, see Appendix C for the collar setups at the different sites.

**Long-term measurements**

For the long term measurements a minimum of 2 (one over the STU and one control) and maximum of 6 (4 over the STU 2 controls) long-term soil flux chambers were deployed. Long-term soil flux measurements of CO$_2$ were carried out using automated chambers and a closed-transient measurement approach (LI-8100 Automated Soil CO$_2$ Flux System, LI-COR). The flux system consisted of three main components: the CO$_2$ gas analyser (non-dispersive infrared gas analyser) hosted in an analyser control unit (LI-8100A), a multiplexer (LI-8150-8) and multiple long-term chambers (LI-8100-104 and LI-8100-104C) (Fig. 3.17 and 3.20). An Ultra-Portable Greenhouse Gas Analyser (Ultra-Portable GGA, model 915-0011, Los Gatos Research), was integrated into the LI-8100 system to measure CH$_4$ fluxes.

The basic concept of the measurement is that chamber is moved by a non-flexible arm over a collar which is inserted into the soil creating a gas loop between the chamber and the analyser to be able to measure the change in gas concentrations under the chamber.
Discrete survey GHG measurements For the long-term measurements, the long-term chambers were rotated between the sites and between the collars. However, this methodology may act to give an incoherent temporal coverage of measurements on each collars and so discrete flux measurements were also carried out manually whist on site over all collars using a portable a survey chamber (see Figure 3.17 and 3.21).

A pressure/vacuum air flow system expands and contracts a bellows to raise and lower the chambers over a soil collar to make the flux measurement.
3.2.3 GHG from vent system

The GHG fluxes were measured from venting system outlets at KM and CC sites at end of each trench. The venting systems were sealed to ensure that the normal air flow was not disturbed. A sampling device (Vent Wizard 800) was constructed from a PVC slip cap and a threaded pipe adapted to fit the cleanout ports, see Figure 3.22a. One port was placed in the slip cap to allow for gas sampling, this port was connected to the UGGA analyser. Air velocity and temperature were also measured in the venting system using a hotwire anemometer (LU8050, TQC Sheen, The Netherlands), see Appendix B.2.
3. SITES AND INSTRUMENTATION

3.2.4 Gas sampling

Additional gas samples were collected from the air stream of the flux measurements integrating a Tee-fitting with a septum (8100-664 Trace Gas Sample Kit) in the gas loop, as shown on Figure 3.23. Four consecutive gas samples were collected during a period of 6-9 minutes using a 50 ml plastic syringe with a needle to withdraw gas and injecting into sealed and pre-evacuated gas vials (20 ml mL GC vials). The CH$_4$ and N$_2$O concentrations were subsequently analysed with a gas chromatography in a laboratory within maximum 72 hours, see Section 3.2.5.
3.2.5 Gas chromatography

Gas samples collected on sites (see Section 3.2.4) were analysed for CH$_4$ and N$_2$O in maximum 72 hours using a Perkin Elmer Clarus 500 gas chromatograph (Perkin Elmer, Waltham, MA, USA) equipped with capillary columns (Elite-Plot Q), flame ionisation detector (FID) and electron capture detectors (ECD), see Table A.1. A headspace autosampler (HS 40, Perkin Elmer) fed the gas samples into the GC systems. The gases were split to the two different detectors and separated in two Elite-Plot Q columns (length, 30 m; inner diameter, 0.53 mm; Perkin Elmer). The FID equipped with a methaniser uses a H$_2$ and air flame to generate ions from the combustion of an organic compound and detector collects and measures the concentration of these ions to determine the amount of compound in the sample. The ECD contains an $^{63}$Ni radioactive foil coating and works with nitrogen carrier.

The gas samples prior to the analysis were pressurised to laboratory atmospheric pressure by releasing the excess pressure with a needle.
3.3 Meteorological monitoring

Meteorological data at KB site were not measured. For this site, hourly rainfall, wind speed and wind direction were recorded 18 km away (Mullingar, N53°53′, W7°35′) by the Irish Meteorological Service.

Meteorological data on all other three sites were collected hourly using automated weather stations (Campbell Scientific) located on sites, equipped with a CR1000 data logger (8DF, 3VX, 2PC, 8DI, 4SDI12, 1SW12V with 2x AM16/32b multiplexer (16DF each) and CF100 compact flash card module) that recorded air temperature $T_{\text{mean}}$, barometric pressure $p$, net radiation $R_{\text{net}}$, rainfall $r$, relative humidity $U$, wind direction $\vec{w}$, and wind speed at $u_2$ 2m height. Combined temperature-humidity probes were mounted in an enclosed radiation shield to protect the instrument from direct exposure to solar heating; net radiometers were mounted on a cross arm facing south to minimize shadowing effects. Mains powered with 7Ah backup battery. All weather stations were located centrally within the STU, 5m from the inlet. All meteorological data were collected in hourly time steps; selected parameters were collected in 10-minute and one-minute time steps (see Appendix B.3).
Analytical Methods

4.1 Chamber flux calculations

4.1.1 Measurements on-site

For CO₂ and CH₄ concentrations recorded by LiCor and/or Los Gatos analysers, the CO₂ and CH₄ fluxes were calculated using SoilFluxPro 4.0 software (LI-COR Biosciences, Inc.). The following method was used for every flux measurement with small alterations depending on whether the flux measurements were over the ST, STU or vent system or the survey or long-term chamber was used.

1. A data file (.81X File extension) recorded by the LI-COR system was opened with the software.

2. All interrupted measurements were discarded.

3. When applicable, the data file (.txt) recorded by the Los Gatos was imported into software with the appropriate time difference.

4. The start and end time of the flux analysis was set separately for CO₂ (30 s and 120 s) and for CH₄ (50 s and 350 s) in order to avoid under or overestimation of fluxes due to the different levels of fluxes and their different gas diffusion in the soil, see Figure 4.1.

5. Linear and exponential lines were fit to gas concentrations versus time by the software.
Figure 4.1: Soil Flux Pro user surface with an example of the flux measurements and calculation.

6. $R^2$-value of the regressions were used as quality control and either the linear or the exponential fit was used depending on the higher $R^2$-value ($CO_2 R^2 < 0.9$, $CH_4 R^2 < 0.8$). Measurements not matching this quality control were discarded from further analysis. To note, when the gas concentration (only for $CH_4$) decreased by time only linear fit was calculated.

7. Negative $CO_2$ flux measurements were excluded. The negative fluxes only occurred during cold, windless nights when the prepurge period was not sufficient to clear out the accumulated $CO_2$ in the dome of the chamber, thereby causing invalid measurements.

8. Potential $CH_4$ flux measurements were also excluded, where the initial $CH_4$ concentration was exceptional high (significantly exceeding the average atmospheric $CH_4$ concentration, above 250 ppb) and resulting unrealistic high $CH_4$ uptake.

9. The fluxes were calculated based on a mass balance approach

$$F = \frac{V_{sys} p_0 (1 - \chi_w)}{sRT_0} \cdot \frac{\partial \chi_c(t)}{\partial t} \quad (4.1)$$

using the total system volume $V_{sys}$ (0.06 m$^3$), the atmospheric pressure at the beginning of the measurement $p_0$ (Pa), the chamber air water vapour mole fraction $\chi_w$ (mol mol$^{-1}$), the soil collar surface area $s$ (0.032 m$^2$), the universal gas constant $R$
4.1. CHAMBER FLUX CALCULATIONS

(8.314 Pa m³ K⁻¹ mol⁻¹), the absolute temperature at the beginning of the measurement $T_0$ (K), and the initial change of chamber water vapour corrected CO₂ mole fraction $\partial \chi_c/\partial t$ (μmol mol⁻¹ s⁻¹) (LI-COR, 2012).

10. All flux values were exported as a .txt file.

11. All further data analysis and statistical analysis have being performed with R (R Core Team, 2014).

4.1.2 Manual gas samples

For the manual gas samples the fluxes were calculated in Excel using the following method.

1. The area of the peaks representative for each gas had a linear relationship with the concentration. The output of gas chromatograph; the peak areas were converted to concentration with known standards. Three standard gas mixes (2, 5 and 10 ppm CH₄ and 200, 500 and 1000 ppb N₂O) were measured at each GC run. The calibration curve varied for CH₄ between 0.00057 and 0.000689; for N₂O between 0.01345 and 0.01945.

2. Data points were discarded if the GC measurements had been interrupted or had malfunctioned.

3. For each flux measurement event, linear regression was used to fit the four measured concentrations (including initial value) versus time.

4. R²-value of the linear regression was used as quality control (R² < 0.7).

5. The fluxes were calculated based on a mass balance approach

$$F = \frac{V_{sys} p_0 (1 - \chi_w)}{sRT_0} \cdot \frac{\partial \chi_c(t)}{\partial t}$$

(4.2)

using the total system volume $V_{sys}$ (0.06 m³), the atmospheric pressure at the beginning of the measurement $p_0$ (Pa), the chamber air water vapour mole fraction $\chi_w$ (mol mol⁻¹), the soil collar surface area $s$ (0.032 m²), the universal gas constant R (8.314 Pa m³ K⁻¹ mol⁻¹), the absolute temperature at the beginning of the measurement $T_0$ (K), and the initial change of chamber water vapor corrected CO₂ mole fraction $\partial \chi_c/\partial t$ (μmol mol⁻¹ s⁻¹) (LI-COR, 2012).

6. All further data analysis and statistical analysis have being performed with R (R Core Team, 2014).
4.1.3 Temperature correction of flux measurements

Some of the temperature sensors had unreliable readings during deployments due to the failure of thermistor inside the chamber, since the accurate flux estimation depends on good chamber temperature reading, these flux values needed to be corrected using the following relationship:

\[
F_{\text{corr}} = \frac{F (T_{\text{cham}} + 273.15)}{T_{\text{corr}} + 273.15}
\]  
(4.3)

where \(F_c\) is obtained flux \(T_{\text{cham}}\) is the bad chamber temperature. \(T_{\text{corr}}\) is the right temperature from the weather station.

4.2 GHG emissions

To compare obtained flux values with previous studies, \(F_c\) was converted to per capita mass emission rates \(E_{\text{cap}}\) [g CO\(_2\) cap\(^{-1}\) d\(^{-1}\)], as follows:

\[
E_{\text{cap}} = \frac{A_{\text{soak}} M_{\text{CO}_2}}{n} \cdot F_c
\]  
(4.4)

assuming a spatially uniform flux distribution and using the number of occupants \(n\) in the household, the surface area of the soakaway \(A_{\text{soak}}\), and the molar mass \(M_{\text{CO}_2}\) of CO\(_2\) [44.01 g mol\(^{-1}\)] as normalization factors.

Fluxes measured from the control lawn were scaled using an area equivalent to the STU (also taking an average of the control fluxes).

4.3 Auxiliary measurement methods

4.3.1 ECH2O Calibration

The calibration used for ECH2O measurements are shown in Figure 4.2. These data were collected in four different soils ranging from silt loam to sand with electrical conductivities ranging up to 5 dSm\(^{-1}\).
4.3.2 Converting bulk EC to pore EC

EC of the solution contained in the soil pores (σp) is a good indicator of the solute concentration in the soil. Extracting pore water from the soil and measuring σp directly is a time-consuming and labour-intensive process.

The ECH2O GS3 sensor measures the EC of the bulk soil surrounding the sensors (σb). Hilhorst (2000) established a linear relationship between the soil bulk dielectric permittivity (ϵb) and σb to derive the pore water EC ϵp [dS m⁻¹]:

\[ σ_p = \frac{ϵ_p \sigma_b}{ϵ_b - ϵ_{σb=0}} \]  

(4.5)

using the real portion of the dielectric permittivity of the soil pore water (ϵp, unitless), bulk EC (σb, [dS m⁻¹]), the real portion of the dielectric permittivity of the bulk soil (ϵb, unitless) and the real portion of the dielectric permittivity of the soil when bulk EC is 0 (ϵ_{σb=0}, unitless).

ϵp can be calculated from soil moisture using a simple formula:

\[ ϵ_p = 80.3 - 0.37(T_{soil} - 20) \]  

(4.6)

where T_{soil} is the soil temperature [°C].

ϵ_{σb=0} is an offset term loosely representing the dielectric of the dry soil. Hilhorst recommends using 4.1 as a generic offset, but in the latest manual it is suggested using 6 as offset.
4. ANALYTICAL METHODS
CO₂ emissions from a septic tank soakaway in a northern maritime climate

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Figure 5.1: Schematic drawing of the septic system with estimated CO₂ emissions from soakaway and control area.
5.1 Abstract

Here, we present the first attempt to quantify long-term and diurnal variations of CO$_2$ fluxes from a soakaway of an on-site wastewater treatment system serving a single house located in a northern maritime climate (Ireland). An automated soil gas flux chamber system was deployed semi-continuously over a period of 17 months, recording hourly flux measurements from the soakaway (F$_{soak}$) and a control site (F$_{control}$). Soil gas fluxes expressed seasonal and diurnal variations: F$_{soak}$ and F$_{control}$ ranged from 0.43μmol CO$_2$ m$^{-2}$ s$^{-1}$ to 100.26μmol CO$_2$ m$^{-2}$ s$^{-1}$ and 0.45μmol CO$_2$ m$^{-2}$ s$^{-1}$ to 19.92μmol CO$_2$ m$^{-2}$ s$^{-1}$ with median fluxes of 6.86μmol CO$_2$ m$^{-2}$ s$^{-1}$ and 5.05μmol CO$_2$ m$^{-2}$ s$^{-1}$, respectively. While temperature, soil water content, and atmospheric pressure were identified as the most significant environmental factors correlated to the release of CO$_2$ from the control site, fluxes from the soakaway showed weaker correlations in regard to environmental factors. Assuming homogeneous spatial flux distributions, the soakaway emitted 15.0 kg yr$^{-1}$ more CO$_2$ into the atmosphere in total compared to a similarly sized control site.

Keywords: anthropogenic greenhouse gas emissions, Ireland, on-site wastewater treatment system, sanitation, soil flux chamber

5.2 Introduction

On-site septic tank systems are a common choice for treating domestic wastewater in areas not connected to a centralized or decentralized wastewater treatment system. In the European Economic Area (EEA) a total of 23% of households are estimated to use on-site wastewater treatment and disposal (EEA, 2013). In particular, regions with a relatively low population density and a significant share of dispersed settlements tend to rely more on on-site septic systems as means for domestic wastewater treatment and disposal; e.g. in Ireland, where 38% of the population lives in rural areas, nearly 30% of households treat their wastewater on site (CSO, 2011). In the Nordic Countries about 34% of the population is connected to an on-site treatment or collection facility (Norin and Tidestrom, 2003) while in the United States about one fifth of the population relies on septic systems (USEPA, 2011).

Globally, there has been a shift from regarding sanitation infrastructure merely as a service for the provision of basic needs towards a more comprehensive implementation and promotion of long-term environmentally sustainable decentralized and on-site treatment
5.2. INTRODUCTION

systems (Massoud et al., 2009; Rosenqvist et al., 2016), particularly in regions currently underserved with basic services provision (Libralato et al., 2012; Parkinson and Tayler, 2003). Currently, with 64% of the urban population in low- and middle-income countries using on-site systems (Hawkins et al., 2014) and an estimated total of 2.4 billion people still lacking access to basic sanitation services globally (UNICEF/WHO, 2015) the total number of installed septic system is likely to rise in the future.

A conventional domestic septic system consists of two components; a septic tank (ST) and a soil dispersal system (SDS). The ST facilitates the initial collection and storage of the raw sewage from one or several households and allows the retention of settleable solids as sludge at the bottom of the tank and flocculent waste as a floating scum layer. While the settled solids are partially anaerobically digested within the tank, the effluent is ideally discharged into the vadose zone via an engineered SDS (e.g. soakaways, percolation trenches/leach fields, mound soil systems, drip line systems).

In Ireland, septic tank systems installed before 1991 had their effluent released into the soil mainly via soakaways (pits back-filled with stone or rubble for effluent disposal). In 1991, the National Standards Authority of Ireland recommended the construction of septic tank systems with larger percolation areas where the effluent would be distributed through percolation trenches – also known as leach fields, drainfields, or infiltration areas. However, of the current more than 450,000 installed on-site systems in Ireland, it is estimated that still approximately 65% remain constructed before the implementation of these revised guidelines (CSO, 2011).

Microbial processes in the ST mainly follow anaerobic pathways for the degradation of organic matter via hydrolysis, acidogenesis, and methanogenesis resulting in the production of CH₄ and CO₂ gas. In the SDS a clogging zone forms at the infiltrative surface for the liquid ST effluent over time. Initial clogging occurs due to an accumulation of suspended solids, organic matter, and chemical precipitation resulting in potentially saturated conditions and ponding of effluent at the infiltrative surface (Beach and McCray, 2003). The increasing impedance to flow, in turn, allows for the formation of a mature biological clogging zone – also known as a microbial biomat – providing significant treatment and attenuation of contaminants before the wastewater reaches the underlying groundwater aquifer (Gill et al., 2007). Bacteria forming the biomat utilize an efficient defense mechanism, producing extracellular polymeric substances (EPS) to create anaerobic microenvironments to protect their bacterial cells. EPS have been characterized as containing significant concentrations of humic substances and polysaccharides and can cause soil clogging leading to lower infiltration rates (McKinley and Siegrist, 2010). Site specific parameters such as system design, mineral subsoil composition, subsoil permeabil-
ity, hydraulic and organic loading rates, and further environmental factors such as soil
temperature and rainfall patterns influence the extend and microbial composition of the
clogging zone ([Beach and McCray, 2003; Gill et al., 2009; Winstanley and Fowler, 2013]).
A continuously fed biomat acts as a potential source of CO$_2$, CH$_4$, and N$_2$O emissions
from transformation and degradation of organic and inorganic contaminants in the soil.

Considering the large number of on-site septic systems in use internationally, potentially
constituting a significant source of GHG emissions, there has been a surprising lack of
direct field measurements of these fluxes to the atmosphere. Most of the existing septic
system emission models rely on load-based calculations or estimated emission factors.
The IPCC provides guidelines on national GHG inventories following an organic load-
based approach to estimate septic system emissions ([IPCC, 2006]). These guidelines only
consider CH$_4$ emissions from anaerobic degradation in STs, disregarding the potential
emissions from microbial degradation processes in the SDS. Direct CO$_2$ emissions from
septic systems are omitted in the GHG inventories as they are of biogenic origin.

Numerous recent studies on septic systems mainly focused on the attenuation of chemical
and biological pollutants and the risk for contamination of groundwater ([Gill et al., 2007;
Godfrey et al., 2007; Katz et al., 2010; Keegan et al., 2014]), wells ([Hynds et al., 2013; Lu
et al., 2008), or surface waters ([Dubber et al., 2016; Ockenden et al., 2014; Rosario et al.,
2014; Withers et al., 2012]) from septic systems. However, there is a limited number of
studies with a scope on quantifying gas emissions from septic systems. Kinnicutt et al.
(1910) reported gas emissions of 39 L m$^{-3}$ treated sewage, of which 75.2% were CH$_4$ and
5.9% were CO$_2$, from a closed municipal septic tank in Worcester (MA, USA) fed with
sewage from domestic and industrial sources. More recent studies by Leverenz et al. (2010)
and Diaz-Valbuena et al. (2011) identified, for the first time, emissions of all three major
GHGs (CH$_4$, CO$_2$, and N$_2$O) from eight septic systems and two SDSs in CA, USA. The
studies noted that the septic tank itself would be the primary source of CH$_4$ emissions
while most of the CO$_2$ is emitted from the SDS with negligible overall N$_2$O emissions.
Emissions from direct flux measurements over the SDSs were deemed negligible. In the
latest study, Truhlar et al. (2016) quantified CH$_4$, CO$_2$, and N$_2$O emissions from SDSs,
sand filters, and vents for a period of three months at eight septic systems in NY, USA.
While the majority of GHG emissions escaped through the roof vent, interpreted as proxy
for direct emissions from the ST surface itself, the SDS was found to be a negligible source
of CO$_2$ but potentially releases N$_2$O.

The existing studies on soil gas fluxes from septic systems have a limited temporal (Septem-
ber to December 2009 in Diaz-Valbuena et al. (2011), June to August 2014 in Truhlar et al.
and spatial span, thus, not being able to fully capture the expected seasonal variability of the emission rates.

Here, we are presenting the first attempt of measuring the CO₂ soil flux from a soakaway and a control area semi-continuously using an automated in-situ flux chamber measurement technique with hourly measurements over a 17-month period. The objective of this study was (i) to quantify the CO₂ emissions of a soakaway receiving domestic septic tank effluent under a normal load in order to detect potential seasonal and diurnal variations, and (ii) to identify potential environmental factors that drive the release of CO₂ over such a system.

5.3 Materials and methods

5.3.1 Study site

CO₂ soil flux measurements were conducted at an on-site wastewater treatment system receiving effluent from a single house in Co. Westmeath, Ireland (N53°24′ W7°30′). The system, consisting of a single-chamber septic tank and subsequent soakaway, was constructed more than 20 years ago. The septic tank has a total capacity of 2.6 m³ with a theoretical hydraulic retention time of 7 d and is fed by a single gravity flow effluent pipe from the household with a fluctuating number of occupants averaging 2. The septic tank was last desludged 3 years before the start of this study. The subsequent soakaway distributes an average 360 l d⁻¹ of effluent over a total area of approximately 6 m².

Co. Westmeath lies in central Ireland and its climate is classified as maritime Cfb (warm temperate, fully humid, warm summer) after Köppen-Geiger (Kottek et al., 2006). The mean annual temperature is 10 °C with mean seasonal minimum and maximum between 4 °C and 15 °C in winter and summer, respectively; average annual rainfall is 1200 mm without significant seasonal variation and 1250 h total annual sunshine (Walsh, 2012). The research site is a homogeneously vegetated grassland previously used for cattle grazing lying in an area of high groundwater vulnerability and is characterized by high permeability, gravelly SAND subsoil according to British Standard classification (BSI, 1999), consisting of till derived mainly from limestone.

The soakaway had previously been the subject of an intensive study which aimed to delineate the effluent plume and determine pollutant attenuation (Gill et al., 2015). It had been instrumented with soil moisture samplers (suction cup lysimeters) throughout its depth. The effluent from the septic tank was found to move rapidly through the
subsoil in a predominantly vertical direction following the overall gradient of the site. Falling-head percolation tests yielded a field saturated conductivity $K_{fs}$ of 1.39 m d$^{-1}$. There was little evidence of the effluent plume extending laterally (apart from along the topographic gradient), consistent with the free draining subsoil characteristics. Despite the high permeability of the site, most of the organic and phosphorus concentration in the effluent was reduced within the biologically active zone of the soakaway which develops at the infiltrative surface over time. Equally, the unsaturated conditions in the soil gave rise to high levels of nitrification which occurred rapidly within the receiving subsoil, with 98% of the inorganic-N present as NO$_3$–N indicating a potential source of N$_2$O emissions (Gill et al., 2015).

5.3.2 Soil gas flux measurement

Long-term soil gas flux measurements of CO$_2$ were carried out using an automated cylindrical chamber system (LI-8100A Automated Soil Gas Flux System, LI-COR Biosciences, Inc.) consisting of a non-dispersive infrared gas analyzer, a multiplexer, and two automated opaque long-term chambers (LI8100-104, LI-COR Biosciences, Inc.).

For each measurement cycle, the chambers were automatically moved over and lowered onto a PVC soil collar (317.8 cm$^2$) permanently inserted approximately 5 cm into the soil to prevent lateral diffusion of CO$_2$ and thus, creating a closed gas loop between chamber and analyzer in order to monitor the change of CO$_2$ concentration inside the chamber (Hutchinson and Livingston, 2001; LI-COR, 2012). Atmospheric pressure ($p_{air}$) and temperature ($T_{air}$) were recorded continuously by the soil gas flux system during the measurements.

The system was deployed semi-continuously between February 2015 and June 2016 with hourly measurements using a 30 s dead band and a 3 min measurement period, i.e. the soil surrounded by the collars was exposed to environmental conditions for 94% of the time. The chambers were operated in sequence with one chamber placed over the soakaway approximately 2 m downslope from the septic tank outlet in an area identified by the previous monitoring with suction cup lysimeters to be within the effluent plume and the other chamber located several metres upslope of the septic tank as control. The locations of the collars were not changed between deployments. The initial 24 h of measurement after installing the soil collars and the initial 6 h of measurement after installing the chambers for each deployment period were discarded to eliminate effects of potential soil disturbance during installation (Bahn et al., 2009).
5.3. MATERIALS AND METHODS

5.3.3 Flux estimation using non-linear regression

Soil CO₂ fluxes $F$ [µmol CO₂ m⁻² s⁻¹] were calculated using SoilFluxPro 4.0 (LI-COR Biosciences, Inc.), which implements a mass balance approach as

\[
F = \frac{V_{\text{cham}} p_0 (1 - \chi_w)}{R s T_0} \cdot \frac{\partial \chi_c(t)}{\partial t} \tag{5.1}
\]

using the volume of the chamber $V_{\text{cham}}$ [m³], the atmospheric pressure at the beginning of the measurement $p_0$ [Pa], the chamber air water vapor mole fraction $\chi_w$ [mol mol⁻¹], the universal gas constant $R$ [Pa m³ K⁻¹ mol⁻¹], the soil collar surface area $s$ [m²], the absolute temperature at the beginning of the measurement $T_0$ [K], and the initial change of chamber water vapor corrected CO₂ mole fraction $\partial \chi_c/\partial t$ [µmol mol⁻¹ s⁻¹] (LI-COR, 2012). $\chi_c(t)$ was calculated using an empirical exponential regression model which is fit to the measured CO₂ concentration data

\[
\chi_c(t) = (\chi_0 - \chi_x) \exp[-a(t - t_0)] + \chi_x \tag{5.2}
\]

with the initial water vapor corrected CO₂ mole fraction $\chi_0$ [mol mol⁻¹], and fit parameters $\chi_x$ [mol mol⁻¹] and $a$ [s⁻¹] (LI-COR, 2012). The fit is used to derive the initial change of chamber water vapor corrected CO₂ mole fraction as the slope of the fit at the time of chamber closure. A non-linear model was chosen in order to reduce the influence of chamber feedback due to increasing resistance to naturally occurring diffusion as the main driver for transport of CO₂ from the soil to the atmosphere following the increase of CO₂ mole fraction inside the chamber during measurement (Davidson et al., 2002; Healy et al., 1996; Kutzbach et al., 2007). For each measurement, the $R^2$-value of the regressions was used as a quality control parameter and measurements with $R^2 < 0.9$ were rejected (less than 0.5% of total measurements).

To compare obtained flux values with previous studies, $F_c$ was converted to per capita mass emission rates $E_{\text{cap}}$ [g CO₂ cap⁻¹ d⁻¹]

\[
E_{\text{cap}} = \frac{A_{\text{soak}} M_{\text{CO}_2}}{n} \cdot F_c \tag{5.3}
\]

assuming a spatially uniform flux distribution and using the number of occupants $n$ in the household, the surface area of the soakaway $A_{\text{soak}}$, and the molar mass $M_{\text{CO}_2}$ of CO₂ [44.01 g mol⁻¹] as normalization factors.
5.3.4 Environmental parameters

During each measurement, air temperature, atmospheric pressure, soil temperature at the surface layer adjacent to the chamber, and volumetric water content of the soil (Soil Moisture Probe EC-5, Decagon Devices, Inc.) at the surface layer (installation depth: 5 cm below soil surface) adjacent to the chamber were recorded. A tipping bucket rain gauge (Casella CEL, Inc.) was deployed from July 2015 until June 2016. Other meteorological parameters (wind speed and direction, relative humidity, and atmospheric pressure) were obtained from a weather station operated by the Irish Meteorological Service which was located 18 km away from the study site in Mullingar (N53°31′ W7°21′) (Met Eireann, 2018). The R package insol (Corripio, 2014) was used to compute daylight hours for the given latitude and dates during data processing.

5.3.5 Statistical analysis

All statistical analysis was performed with R version 3.2.3 (R Core Team, 2014). Welch’s unequal variances t-tests were used to address the significance of whether observed CO₂ fluxes from the soakaway \( F_{\text{soak}} \) were different from the fluxes observed at the control site \( F_{\text{control}} \). Relationships between CO₂ fluxes and environmental variables were analyzed with multiple linear regression models; the relative importance of the predictors was calculated using the LMG method with bootstrap confidence intervals \( (b = 1000) \) and an averaging of the sequential sum-of-squares obtained from all possible orderings of the predictors, using the R package relaimpo (Gromping, 2006). All variables \( x \) were examined for evidence of normality and homogeneity and log-transformed \( [\lg (x + 1)] \) if needed to attain normality.

5.4 Results and Discussions

5.4.1 Temporal variation

CO₂ fluxes were measured over the soakaway \( F_{\text{soak}} \) and control site \( F_{\text{control}} \) semi-continuously, including 3709 hourly paired measurements. \( F_{\text{soak}} \) and \( F_{\text{control}} \) ranged from 0.43 to 100.26 μmol CO₂ m⁻² s⁻¹ and 0.45 to 19.92 μmol CO₂ m⁻² s⁻¹ with medians of 6.86 μmol CO₂ m⁻² s⁻¹ and 5.05 μmol CO₂ m⁻² s⁻¹, respectively. The normalized total yearly mean emissions over the soakaway were 57.1 kg CO₂ compared to 42.1 kg CO₂ for the undisturbed control, yielding net emissions of 15.0 kg CO₂ yr⁻¹.
Overall, the calculated flux values showed a bi-modal distribution for both $F_{\text{soak}}$ and $F_{\text{control}}$. Soakaway fluxes up to 20-times higher than the overall median were observed during periods of extremely high fluxes in October 2015 (Figure 5.2A). Over the control site the bi-modal distribution was controlled by soil temperature with an identified threshold of $10^\circ\text{C}$ (Figure 5.3). In order to ensure normal distribution of flux values for statistical analysis, data were therefore divided into four subgroups according to temporal (October and non-October fluxes) and temperature-related ($T_{\text{soil}} < 10^\circ\text{C}$ and $T_{\text{soil}} \geq 10^\circ\text{C}$) selection rules. Despite seasonal variations in soil gas fluxes resulting in lower median fluxes from the soakaway compared to the control in April 2015, July 2015, and May 2016 (Figure 5.2A), Welch’s two-sample t-tests for unequal variances confirmed that, overall, the mean soakaway fluxes were significantly higher than fluxes observed over the control site for each subgroup (Table 5.2).

Day-night soil gas flux variations (Figure 5.2B) were observed with higher total median fluxes during day time (soakaway: 7.62 $\mu$mol CO$_2$ m$^{-2}$ s$^{-1}$; control: 5.77 $\mu$mol CO$_2$ m$^{-2}$ s$^{-1}$) compared to night time (soakaway: 6.02 $\mu$mol CO$_2$ m$^{-2}$ s$^{-1}$; control: 4.38 $\mu$mol CO$_2$ m$^{-2}$ s$^{-1}$). Welch’s two-sample t-tests identified significantly higher mean day time fluxes in three of four subgroups (control fluxes at soil temperatures above and below 10$^\circ\text{C}$, and non-October soakaway fluxes), while during the October measurements over the soakaway mean day time fluxes were lower than mean night time fluxes (Table 5.2). Hence, monitoring soakaway fluxes during day time only could lead to potential overestimation of total emissions.

Comparing the diurnal pattern of soakaway and control soil gas fluxes, the median fluxes from the soakaway expressed, on average, an earlier increase and decrease starting from around 9 a.m. and 3 p.m. respectively (Figure 5.2C). The earlier increase in hourly median flux from the soakaway is presumed to be due to the morning use of water from the household resulting in partially treated wastewater from the ST to be discharge into the soakaway.

Table 5.1: Statistics for comparing soakaway and control soil gas fluxes using Welch’s two-sample t-tests for unequal variances

<table>
<thead>
<tr>
<th></th>
<th>soakaway$^a$</th>
<th>control$^a$</th>
<th>t</th>
<th>df</th>
<th>p</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>mean  SD</td>
<td>mean  SD</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>non-October, $T_{\text{soil}} &lt; 10^\circ\text{C}$</td>
<td>5.17  3.69</td>
<td>3.00  1.35</td>
<td>20.99</td>
<td>1837</td>
<td>&lt;0.05</td>
</tr>
<tr>
<td>non-October, $T_{\text{soil}} \geq 10^\circ\text{C}$</td>
<td>8.00  3.20</td>
<td>7.33  2.66</td>
<td>6.69</td>
<td>3330</td>
<td>&lt;0.05</td>
</tr>
<tr>
<td>October, $T_{\text{soil}} &lt; 10^\circ\text{C}$</td>
<td>36.52 10.94</td>
<td>4.13  0.72</td>
<td>11.44</td>
<td>14</td>
<td>&lt;0.05</td>
</tr>
<tr>
<td>October, $T_{\text{soil}} \geq 10^\circ\text{C}$</td>
<td>53.85 19.62</td>
<td>5.14  0.93</td>
<td>56.60</td>
<td>522</td>
<td>&lt;0.05</td>
</tr>
</tbody>
</table>

$^a$ fluxes in $\mu$mol CO$_2$ m$^{-2}$ s$^{-1}$; SD = standard deviation, t = test statistic, df = degrees of freedom, p = p-value
5. \( \text{CO}_2 \) EMISSIONS FROM SEPTIC TANK SOAKAWAY

Table 5.2: Statistics for comparing day and night time soil gas fluxes using Welch’s two-sample t-tests for unequal variances

<table>
<thead>
<tr>
<th></th>
<th>day time(^a)</th>
<th>night time(^a)</th>
<th>t</th>
<th>df</th>
<th>p</th>
</tr>
</thead>
<tbody>
<tr>
<td>soakaway, non-October</td>
<td>7.37 3.73</td>
<td>5.89 3.52</td>
<td>11.50</td>
<td>3098</td>
<td>&lt;0.05</td>
</tr>
<tr>
<td>soakaway, October</td>
<td>51.14 18.72</td>
<td>55.07 20.18</td>
<td>-2.33</td>
<td>516</td>
<td>&lt;0.05</td>
</tr>
<tr>
<td>control, ( T_{\text{soil}} &lt; 10 \degree \text{C} )</td>
<td>3.09 1.51</td>
<td>2.96 1.23</td>
<td>1.85</td>
<td>1152</td>
<td>&lt;0.07</td>
</tr>
<tr>
<td>control, ( T_{\text{soil}} \geq 10 \degree \text{C} )</td>
<td>7.41 2.76</td>
<td>5.89 1.81</td>
<td>15.68</td>
<td>2238</td>
<td>&lt;0.05</td>
</tr>
</tbody>
</table>

\(^a\) fluxes in \( \mu\text{mol CO}_2 \text{ m}^{-2} \text{s}^{-1} \); SD = standard deviation, t = test statistic, df = degrees of freedom, p = p-value

Figure 5.2: The monthly (A), diurnal (B), and hourly (C) variation of log-scaled \( \text{CO}_2 \) fluxes from soakaway (white) and control (grey) as density curve bean plots with median values (black lines) for each group. Dashed lines represent median soil gas flux values from the soakaway (black) and control (grey).
5.4. RESULTS AND DISCUSSIONS

5.4.2 Impact of meteorological factors on carbon dioxide flux

Multilinear regression models were applied to the log-scaled flux data using the environmental parameters air temperature $T_{\text{air}}$, atmospheric pressure $p_{\text{air}}$, relative humidity $\phi_{\text{rel}}$, water vapor corrected CO$_2$ mole fraction $\chi_c$, soil temperature $T_{\text{soil}}$, soil moisture $\theta_{\text{soil}}$, wind speed $v_{\text{wind}}$, and wind direction $d_{\text{wind}}$ as continuous and rainfall $P$ as binary input variables. A total of 132 measurements (i.e. 3.5\%) was discarded due to incorrect soil moisture data. October flux data were analyzed separately in order to attain a normal distribution for the data set.

The analysis shows that 33\% and 81\% of the variation in soakaway and control fluxes, respectively, can be explained by environmental parameters (Figure 5.3). The relative importance of the predictors expressed similar patterns for both sites. While air and soil temperature had the highest relative importance, humidity, CO$_2$ mole fraction, wind speed, wind direction, and rain were insignificant. Soil moisture and atmospheric pressure were significant predictors only for the control fluxes.

In October, only 24\% of the variation in fluxes from the soakaway could be explained by environmental predictors and the distribution of their relative importance was distinctly different from the non–October measurements; soil moisture, CO$_2$ mole fraction, and soil temperature express the highest relative importance with relatively large 95\% bootstrap confidence intervals (Figure 5.3). Further regression models were tested for the flux data collected in October, but the environmental parameters recorded in this study were not able to explain the uniquely high fluxes.
5. CO₂ EMISSIONS FROM SEPTIC TANK SOAKAWAY

To better understand the potential drivers of soil gas fluxes from the soakaway and control site, the most important predictors (responsive variance > 5%) were further analyzed for potential correlation using a linear regression model and Pearson correlation coefficient \( r \) (Figure 5.3). Positive correlations were found between fluxes and both air and soil temperature as well as between \( F_{\text{control}} \) and \( p_{\text{air}} \) and a negative correlation was identified between \( F_{\text{control}} \) and \( \theta_{\text{soil}} \). However, no correlation was found for \( F_{\text{soak}} \).

14\% and 25\% of variation in \( F_{\text{soak}} \) can be explained with \( T_{\text{soil}} \) and \( T_{\text{air}} \), respectively. For \( F_{\text{control}} \) this relation is reverse, with 64\% and 42\% of the variation explained by \( T_{\text{soil}} \) and \( T_{\text{air}} \), respectively. The regressions, for both soakaway and control fluxes related to air temperature had similar slopes with \((0.34 \pm 0.01) \text{μmol CO}_2 \text{ m}^{-2} \text{s}^{-1} \text{°C}^{-1}\) and \((0.36 \pm 0.01) \text{μmol CO}_2 \text{ m}^{-2} \text{s}^{-1} \text{°C}^{-1}\), respectively (slope ± SE), resulting in similar flux increases with increasing air temperature. However, both \( F_{\text{soak}} \) and \( F_{\text{control}} \) expressed a higher sensitivity towards increasing soil temperature with \(0.51 \text{μmol CO}_2 \text{ m}^{-2} \text{s}^{-1} \text{°C}^{-1}\) and \(0.80 \text{μmol CO}_2 \text{ m}^{-2} \text{s}^{-1} \text{°C}^{-1}\), respectively. 10\% and 42\% of \( F_{\text{control}} \) variation can be explained with \( p_{\text{air}} \) and \( \theta_{\text{soil}} \), respectively.

The seasonal and diurnal variations of fluxes appeared to mimic the change in air temperature and related soil temperature, as indicated by the high correlation between flux and temperature values. \( F_{\text{soak}} \) expressed a stronger correlation with \( T_{\text{air}} \) than \( T_{\text{soil}} \).

Apart from heavy rainfall events, the soil at the control site is normally unsaturated. However, due to a continuous recharge with partially treated effluent from the ST, the vadose zone surrounding the soakaway is mostly close to or at saturated water content. In future studies, the exact hydraulic and organic loading rates of wastewater discharged into the SDS and vadose zone soil water content should be monitored in order to being able to derive more accurate correlations with non-environmental factors governing the production and release of GHGs from septic systems.

5.4.3 Comparison with previous studies

CO₂ flux rates were converted to per capita mass emission rates \( E_{\text{cap}} \) (Equation 5.3) and compared with previous studies (Table 5.3). The IPCC and USEPA consider CO₂ emissions from wastewater treatment as biogenic and, thus, do not included them into their guidelines for GHG estimations from on-site systems. \cite{Diaz-Valbuena2011} observed significant CO₂ emissions from the septic tank and venting system, and found negligible emissions from the leach field. Mean emissions from this study including all measurement showed good agreement with emission values estimated by \cite{Truhlar2016} who attempted to monitor CO₂ fluxes from SDSs, and as well as from the sand
5.4. RESULTS AND DISCUSSIONS

Figure 5.4: Relative importance of air temperature $T_{air}$, atmospheric pressure $p_{air}$, relative humidity $\phi_{rel}$, water vapor corrected CO$_2$ mole fraction $\chi_c$, soil temperature $T_{soil}$, soil volumetric water content $\theta_{soil}$, wind speed $v_{wind}$, wind direction $d_{wind}$, and rain $P$ for predicting $\lg F_{\text{control}}$ (grey), $\lg F_{\text{soak}}$ (white), and $\lg F_{\text{soak}}$ (dark grey) with 95% bootstrap confidence intervals (method = LMG, $b = 1000$).

filters, and the vents. However, the mean flux value from our study, 155 g CO$_2$ cap$^{-1}$ d$^{-1}$, is highly biased by the extremely high fluxes found in October 2015. Excluding the October data results in mean CO$_2$ emissions rates of 78 g CO$_2$ cap$^{-1}$ d$^{-1}$, compared to the 602 g CO$_2$ cap$^{-1}$ d$^{-1}$ from the October data alone.

5.4.4 Implications of the study

The semi-continuous measurement of CO$_2$ flux from a septic tank soakaway ($F_{\text{soak}}$) and control site ($F_{\text{control}}$) over a period of 17 months expressed seasonal and diurnal variations: $F_{\text{soak}}$ and $F_{\text{control}}$ ranged from 0.43 $\mu$mol CO$_2$ m$^{-2}$ s$^{-1}$ to 100.26 $\mu$mol CO$_2$ m$^{-2}$ s$^{-1}$ and 0.45 $\mu$mol CO$_2$ m$^{-2}$ s$^{-1}$ to 19.92 $\mu$mol CO$_2$ m$^{-2}$ s$^{-1}$ with median fluxes of 6.86 $\mu$mol CO$_2$ m$^{-2}$ s$^{-1}$

Table 5.3: Comparing results from this and previous studies of CO$_2$ emissions from septic systems

<table>
<thead>
<tr>
<th>Study</th>
<th>Emissions [g CO$_2$ cap$^{-1}$ d$^{-1}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diaz-Valbuena et al. (2011)</td>
<td></td>
</tr>
<tr>
<td>septic tank$^a$</td>
<td>33.3 ± 2.7</td>
</tr>
<tr>
<td>vent pipe$^a$</td>
<td>335.0 ± 2.1</td>
</tr>
<tr>
<td>leach field$^a$</td>
<td>negligible</td>
</tr>
<tr>
<td>Truhlar et al. (2016)</td>
<td></td>
</tr>
<tr>
<td>vent pipe$^a$</td>
<td>160.0 ± 3.2</td>
</tr>
<tr>
<td>sand filter$^b$</td>
<td>120 ± 83</td>
</tr>
<tr>
<td>leach field$^b$</td>
<td>130 ± 120</td>
</tr>
<tr>
<td>This study (all measurements)</td>
<td>155 ± 208</td>
</tr>
<tr>
<td>This study (excluding October)</td>
<td>78 ± 42</td>
</tr>
<tr>
<td>This study (only October)</td>
<td>602 ± 222</td>
</tr>
</tbody>
</table>

$^a$ geometric mean ± SD; $^b$ arithmetic mean ± SD.
Figure 5.5: Dependence of measured fluxes ($F_{\text{soak}}$ and $F_{\text{control}}$) on environmental factors (air temperature $T_{\text{air}}$, soil temperature $T_{\text{soil}}$) and of $F_{\text{control}}$ on atmospheric pressure $p_{\text{air}}$ and soil volumetric water content $\theta_{\text{soil}}$. Plots for $F_{\text{soak}}$ are color-coded by $F_{\text{soak}} : F_{\text{control}}$ ratio.
5.4. RESULTS AND DISCUSSIONS

and 5.05 μmol CO₂ m⁻² s⁻¹, respectively. This means, assuming a spatially homogeneous flux distribution, the soakaway emitted a total of 15.0 kg yr⁻¹ more CO₂ into the atmosphere (soakaway emissions: 42.1 kg CO₂ yr⁻¹) compared to a similarly sized control site (control emissions: 57.1 kg CO₂ yr⁻¹).

However, this study was limited with respect to determining the spatial variation of CO₂ flux from the SDS due to the number of soil gas flux chambers available. Thus, it is not possible to conclude whether the fluxes measured over the soakaway were representative of the average total CO₂ fluxes resulting from the main plume of percolating effluent. Especially, the 20-fold higher median emissions observed in October (compared to all other monitoring months) could not be explained with the measured environmental parameters alone and give rise to the questions whether a shifting effluent plume in the vadose zone could result in spatially and temporarily highly variable soil gas flux emissions. It is possible that the area of maximum emissions is moving with the effluent plume and that the October measurements captured this phenomenon.

Future studies should (i) further investigate the spatial distribution of fluxes using a multiple chamber set-up to account for the potentially high spatial variability of GHG fluxes over a soakaway or other SDSs in order to be able to more accurately predict total mass emission rates per unit area from such systems, and (ii) monitor the soil moisture conditions of the system in question more closely and at different depths in order to establish a better understanding of the actual spatial and temporal variation of the effluent plume in the soil.

Among the recorded environmental parameters, atmospheric and soil temperature were the best predictors of CO₂ fluxes from the soakaway. For a more comprehensive understanding of the temporal variations of soil gas fluxes from septic systems, future studies should also record quantitative and qualitative parameters and the spatial distribution of the septic tank effluent released to the SDS.

To understand total septic system emissions, long-term integrated studies for simultaneous monitoring of major GHG emissions (CO₂, CH₄, and N₂O) are needed, including direct measurements on the ST surface and a variation of measuring points over the SDS to accurately capture the spatial and temporal heterogeneity of gas fluxes.
Acknowledgements

This work was supported by the Science Foundation Ireland under grant number 13/IA/1923. The authors thank Lara Brouillet, Ana Alves Cavalcanti, Carolin Kiefer, Michael Swenson, and Patrick Veale for assistance with field work. We are also grateful to the house owner for enabling continued access to the research site.

Author contributions

L.G. conceived the study. C.S., J.K., and L.G. planned the experiments. C.S. and J.K. performed the experiments and analyzed data. C.S., J.K., and L.G. wrote the manuscript. All authors interpreted the results and reviewed the manuscript.
Spatial and temporal variation of CO\textsubscript{2} and CH\textsubscript{4} emissions from a septic tank soakaway

Celia Somlai, Jan Knappe and Laurence Gill

This manuscript was accepted for publication on the 30\textsuperscript{th} of April 2019 to the Science of the Total Environment.

Schematic drawing of the septic system with estimated CO\textsubscript{2} and CH\textsubscript{4} emissions from soakaway and control area.
6.1 Abstract

CO₂ and CH₄ flux measurements over a septic tank soakaway located in a northern maritime climate (Ireland) were conducted for a period of 81 days using a multi-chamber automated soil gas flux chamber system with high spatial and temporal resolution. Overall median CO₂ fluxes were 7.28 and 6.40 µmol CO₂ m⁻² s⁻¹ from the soakaway and control soil, respectively. Overall median CH₄ fluxes were 0.28 and 0.67 nmol CH₄ m⁻² s⁻¹ from the soakaway and control soil, respectively. While CO₂ fluxes expressed strong diurnal variability driven by soil temperature, CH₄ fluxes were less affected by environmental factors and effectively limited to the first few meters from the septic tank. However, localized CH₄ degassing events were observed during drying conditions with up to 60-times higher fluxes compared to the overall median. The soakaway was found to be a net emitter of both CO₂ and CH₄, releasing a total of 7.327 kg CO₂ yr⁻¹ and 0.033 kg CO₂ Eq. yr⁻¹, respectively. The apparent spatio-temporal heterogeneity of observed soil gas fluxes identified in this study emphasizes the importance of integrating measurements with both high spatial and temporal resolution from on-site installations as engineered nature-based solutions.

Keywords: carbon dioxide, methane, anthropogenic greenhouse gas emissions, on-site wastewater treatment system, soil flux chamber

6.2 Introduction

The process of treating domestic wastewater can be a significant source of both direct and indirect anthropogenic greenhouse gas (GHG) emissions (Sweetapple et al., 2014; Paravicini et al., 2016). While the entire wastewater sector is estimated to contribute up to 1.5% of total GHG and 5% of non-CO₂ GHG emission globally, the sector’s contribution to overall waste-related GHG emissions is estimated to increase from 36% in 1990 to 42% by 2030 (Bogner et al., 2008; USEPA, 2012). Most research in this area to date, however, has focused on large-scale, centralized treatment systems (e.g. Czepiel et al., 1995; Johansson et al., 2004; Cakir and Stenstrom, 2005). Despite an estimated 20% of the population relying on on-site wastewater treatment and disposal in both the European Economic Area and the United States (EEA, 2013; USEPA, 2016), only a limited number of studies, so far, has been based on direct measurements of GHG emissions from on-site and decentralized wastewater treatment systems (Leverenz et al., 2010; Diaz-Valbuena et al., 2011; Truhlai et al., 2016; Somlai-Haase et al., 2017). In low- and middle-income countries, the share of these systems is significantly higher than the global average and estimated to provide
improved sanitation for up to 64% of the population (Hawkins et al., 2013). With an ever-growing global population—predominantly in areas currently unserved by centralised sanitation solutions—there is a general imperative to build and improve sanitation infrastructure globally as manifested in the Sustainable Development Goal 6 on clear water and sanitation (United Nations, 2015). Septic systems, commonly consisting of a septic tank for initial collection, storage and partial treatment of domestic wastewater followed usually by some form of soil treatment unit (STU) for additional treatment and disposal into subsurface soil, are environmentally and economically sustainable on-site solutions for wastewater treatment, if properly constructed and managed.

As septic systems rely on naturally occurring biogeochemical processes for wastewater treatment, they are inherently suitable for off-the-grid solutions and in areas with disperse settlement patterns. Despite their apparently simple design, the biogeochemical processes facilitating the treatment of effluent in septic systems are rather complex and involve a wide range of microbial populations and dissolved oxygen concentrations (Beal et al., 2005; Tomaras et al., 2009). Wilhelm et al. (1994) presented a conceptual model describing two main redox environments for on-site systems. The first redox zone in the septic tank is characterised as an anaerobic environment with initially high concentrations of organic C and N, and low dissolved oxygen levels. Here, organic matter is mainly degraded via hydrolysis, acidogenesis, and methanogenesis producing both CO$_2$ and CH$_4$. In the second redox zone—the STU—both aerobic and anaerobic conditions can exist. Gaseous diffusion in the unsaturated zone receiving the partially treated effluent supplies oxygen for aerobic oxidation of organic C resulting in the production of CO$_2$. In the absence of oxygen, e.g. in saturated microsites, ponded infiltration trenches, or within mature biomats at the infiltrative surface, anaerobic organisms such as methanogenic bacteria break down insoluble organic compounds to CO$_2$ and CH$_4$. Research by Fernández-Baca et al. (2018) indicated recently that elevated concentrations of CH$_4$ in the upper soil layer above the infiltration trenches, can sustain a population of CH$_4$ consuming bacteria, which may act to reduce overall net CH$_4$ fluxes from the soil to the atmosphere.

Prevalent septic system designs vary by country and consider local regulations, climatic conditions and final discharge options. In 1991, the National Standards Authority of Ireland (NSAI) implemented guidelines recommending the construction of septic tank systems with percolation areas where the effluent is distributed through a series of parallel trenches providing adequate hydraulic loading of effluent onto the underlying unsaturated soil where the majority of the overall treatment process occurs. However, census data suggests that up to 65% of the existing on-site systems currently in use in the Republic of Ireland were installed before 1991 (CSO, 2011); most of which discharge their effluent to soil via soakaways not meeting the standards set within the updated guidelines. These
soakaways consist mainly of a gravity–flow fed effluent pipe releasing partially treated effluent from the septic tank directly into a pit backfilled with stone and rubble, thus, limiting the application of effluent onto a considerably smaller overall surface area compared to trench–based STUs. The relative prevalence of such soakaway systems in the Republic of Ireland has a wide range of implications for both the hydraulic and pollutant loading onto the receiving soil and renders a significant number of on–site systems vulnerable to becoming sources of groundwater pollution and GHG emissions.

To date, only a limited number of studies tried to quantify direct GHG emissions to the atmosphere from septic systems. Leverenz et al. (2010) and Diaz-Valbuena et al. (2011) measured CH₄, CO₂ and N₂O fluxes from eight conventional septic tanks in California, reporting fluxes of −11, 33.3 and 0.005 g cap⁻¹ d⁻¹, respectively. Additional measurements over two STUs yielded negligible fluxes for all three gases. Truhlar et al. (2016) investigated GHG emissions from STUs (trench–based STUs and sand filters) and the septic tank vent system and found mean CO₂ and N₂O emissions of 130 and 0.022 g cap⁻¹ d⁻¹ as well as a mean CH₄ uptake of 0.0038 g cap⁻¹ d⁻¹ over the STUs. The emissions reported in these studies resulted from a series of respective one–off measurements at several sampling points above the STU, assuming a uniform temporal and spatial distribution of soil gas fluxes. Somlai-Haase et al. (2017) deployed a single soil CO₂ flux chamber for a period of 17 months over a septic tank soakaway to quantify long–term and diurnal CO₂ flux variations and found fluxes ranging from 0.43 μmol CO₂ m⁻² s⁻¹ to 100.26 μmol CO₂ m⁻² s⁻¹ with a median of 6.86 μmol CO₂ m⁻² s⁻¹. While temperature, soil volumetric water content (VWC), and atmospheric pressure were identified as the most significant environmental drivers correlated to the release of CO₂ from natural control soil adjacent to the septic system, fluxes from the soakaway expressed weaker correlations to environmental factors. Assuming a homogeneous spatial flux distribution, the soakaway was found to contribute a net 15.0 kg CO₂ yr⁻¹ into the atmosphere compared to natural soil.

The research presented in this study provides, to our knowledge, the first attempt to integrate continuously quantified emissions of both CO₂ and CH₄ from an STU (in the form of a soakaway) using hourly observations with spatially distributed soil gas flux measurements along a series of sections across the effective percolation area.
6.3 Materials and methods

6.3.1 Study design

Automated long-term CO\textsubscript{2} and CH\textsubscript{4} soil gas flux measurements were conducted semi-continuously between 3\textsuperscript{rd} April and the 7\textsuperscript{th} July 2016 over a septic tank soakaway receiving primary treated effluent from a single house in Co. Westmeath, Ireland. The number of occupants in the house fluctuated over the course of this study, averaging two. Table 6.1 summarizes the characteristics of the study site and wastewater treatment system. A detailed description of the site location, history and prevalent meteorological conditions is given in Somlai-Haase et al. (2017).

Table 6.1: Site and wastewater treatment system characterisation.

<table>
<thead>
<tr>
<th>Site</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Aquifer category</td>
<td>Rkd\textsuperscript{a}</td>
</tr>
<tr>
<td>Groundwater vulnerability</td>
<td>High</td>
</tr>
<tr>
<td>Topsoil</td>
<td>BminDW\textsuperscript{b}</td>
</tr>
<tr>
<td>Subsoil</td>
<td>TLs\textsuperscript{c}</td>
</tr>
<tr>
<td>Area use</td>
<td>Pasture land</td>
</tr>
<tr>
<td>Climate</td>
<td>Cfb\textsuperscript{d}</td>
</tr>
<tr>
<td>Average annual temperature</td>
<td>10 °C</td>
</tr>
<tr>
<td>Average annual rainfall</td>
<td>1200 mm</td>
</tr>
<tr>
<td>Total annual sunshine</td>
<td>1250 h</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>System</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>No. of occupants</td>
<td>2</td>
</tr>
<tr>
<td>Septic tank</td>
<td>Single-chamber</td>
</tr>
<tr>
<td>Capacity (m\textsuperscript{3})</td>
<td>2.6</td>
</tr>
<tr>
<td>Desludged</td>
<td>2012</td>
</tr>
<tr>
<td>HRT\textsuperscript{e}(d)</td>
<td>7.2</td>
</tr>
<tr>
<td>Effluent conveyance</td>
<td>Gravity flow</td>
</tr>
<tr>
<td>STU</td>
<td>Soakaway</td>
</tr>
<tr>
<td>Construction</td>
<td>before 1995</td>
</tr>
<tr>
<td>Area (m\textsuperscript{2})</td>
<td>6</td>
</tr>
<tr>
<td>$K_{fs}$ (m d\textsuperscript{-1})</td>
<td>1.39</td>
</tr>
</tbody>
</table>

\textsuperscript{a} Regionally Important Aquifer - Karstified (diffuse) (GSI, 2018);  
\textsuperscript{b} Deep well drained mineral, mainly basic soil (from Teagasc soil maps, (GSI, 2018));  
\textsuperscript{c} Limestone till subsoil (GSI, 2018);  
\textsuperscript{d} Warm temperate, fully humid, warm summer (Walsh, 2012);  
\textsuperscript{e} Hydraulic Retention Time.
6.3.2 Carbon dioxide and methane flux measurements

Long-term soil gas flux measurements of CO₂ and CH₄ were carried out using an automated system consisting of cylindrical opaque long-term chambers (LI8100-104, LI-COR Biosciences, Inc.), a multiplexer, and a non-dispersive infrared gas analyzer for analysis of CO₂ (LI-8100A Automated Soil Gas Flux System, LI-COR Biosciences, Inc.). A detailed description of the long-term soil gas flux system employed in this study can be found in Somlai-Haase et al. (2017). Contrary to Somlai-Haase et al. (2017), however, a total of 5 chambers were simultaneously deployed at specific sampling locations over the soakaway marked by soil collars made from PVC pipe sections inserted approximately 5 cm into the soil to provide a better understanding of the spatial variation of potential soil gas fluxes, as shown in Figure 6.1. Additionally, a gas analyzer capable of CH₄ detection (Ultraportable Greenhouse Gas Analyzer, model 915-0011, Los Gatos Research) was integrated into the gas loop according to instructions provided by Li-Cor (2015).

The automated soil gas flux system was programmed to take hourly measurements for each chamber using a 6 min incubation period in sequence with a 3 min gas line purging in between measurements. Water vapor corrected gas concentrations for both CO₂ and CH₄ were recorded at 1 s intervals for flux calculations.

Soil gas flux measurements were conducted between 3rd April and 7th July 2016 with two different setups – in setup 1, CO₂ and CH₄ fluxes were continuously measured for 55 days over the soakaway from soil collars 1 to 4 installed at distances of 1, 2, 3 and 4 m from the septic tank on the central axis parallel to the natural gradient on site, respectively, as previous studies tracing the effluent as it percolates through the soil indicated this to be the dominant direction of effluent dispersal in the soil (Gill et al., 2015); for setup 2 the collars located at 2 and 3 m distance were moved by 1 m perpendicular to the natural gradient in opposite directions to location 5 and 6 (see Figure 6.1) to capture potential emissions resulting from a lateral spread of the effluent in the soakaway and CO₂ and CH₄ fluxes were continuously measured for another 26 days. The overall radius of the measurements was determined by visual observation based, primarily, on the presence of indicator vegetation as compared with the surrounding vegetation. Control measurements capturing the natural variation of CO₂ and CH₄ fluxes from soil unaffected by effluent loading were obtained from collar C located 2 m uphill from the septic tank (see Figure 6.1). All equipment deployed in the field was taken to the Environmental Engineering Lab at Trinity College Dublin for cleaning and maintenance in between the two measurement runs. Note, that the positions of collar 2 and C were identical to the ones used for measuring CO₂ emissions over the soakaway and control, respectively, in a previous study (Somlai-Haase et al., 2017).
6.3. MATERIALS AND METHODS

Figure 6.1: Study design with (A) the layout of the study site and treatment system, including soil gas flux chamber positions over the soakaway (collars 1 to 6) and control soil (collar C) and (B) deployment of soil gas flux chambers on collars 1 to 4 during setup 1.

6.3.3 Flux calculation

Soil CO$_2$ and CH$_4$ fluxes were calculated using the SoilFluxPro 4.0 software package (LI-COR Biosciences, Inc.). First, the time series data representing CH$_4$ concentrations from the CH$_4$ analyzer were imported into the software and time stamps were merged with the CO$_2$ data. Then, start and end times of the individual chamber measurements for flux calculations were set separately for CO$_2$ (30 and 120 s, respectively) and CH$_4$ (50 and 350 s, respectively) in order to reflect varying soil gas diffusion rates for the respective gases. Discarding the first 30 and 50 s of CO$_2$ and CH$_4$ concentration measurements, respectively, accounts for potential disturbances caused by chamber closure and the time needed to establish complete mixing of the chamber headspace. Both linear and exponential models were then fit to the gas concentration data. Regression $R^2$-values were used as quality control and selection criterion for using either the linear or exponential model for flux calculations (the model with higher $R^2$-value was selected). Observations resulting in model fits with low overall $R^2$-values ($R^2 < 0.9$ for CO$_2$ and $R^2 < 0.8$ for CH$_4$) were assumed compromised and discarded, representing a total of 0.5 and 34% of CO$_2$ and CH$_4$ measurements, respectively. It has been suggested that flux values estimated by linear models tend to systematically underestimate actual fluxes while exponential models based on diffusion theory result in more accurate flux calculations ([Healy et al., 1996; Nakano et al., 2004; Levy et al., 2011]). Exponential models, though, are not able to capture net uptake phenomena (e.g. for CH$_4$) and are generally less robust compared to linear models. However, Forbrich et al. (2010) observed that while for CH$_4$ non-linear
changes of concentration over time occurred mainly during periods of changing VWC, overall, linear models were the best-fitted model for a majority of CH\textsubscript{4} measurements across sites with a variety of soil VWCs.

In addition, 3\% of the remaining CH\textsubscript{4} flux values were excluded due to recorded initial CH\textsubscript{4} concentrations significantly exceeding the mean atmospheric CH\textsubscript{4} concentration (i.e., \textgreek{i} 250 ppb). These events were observed solely during night time and mainly at sampling location 1 (closest to the septic tank) which also exhibited the highest overall CH\textsubscript{4} emissions. It has been widely reported that night time soil gas flux measurements are prone to be affected by stratified, low-turbulence atmospheric conditions with steep vertical concentration gradients at the soil–atmosphere interface (Schneider et al., 2009; Koskinen et al., 2014; Grres et al., 2016).

Final flux $F$ calculations were based on a mass balance approach

$$F = \frac{V_{sys} p_0 (1 - \chi_w)}{RsT_0} \cdot \frac{\partial \chi_c(t)}{\partial t}$$

using the total system volume $V_{sys}$ (0.06 m\textsuperscript{3}), the atmospheric pressure at the beginning of the measurement $p_0$ (Pa), the chamber air water vapor mole fraction $\chi_w$ (mol mol\textsuperscript{-1}), the universal gas constant $R$ (8.314 Pa m\textsuperscript{3} K\textsuperscript{-1} mol\textsuperscript{-1}), the soil collar surface area $s$ (0.032 m\textsuperscript{2}), the absolute temperature at the beginning of the measurement $T_0$ (K), and the initial change of chamber water vapor corrected CO\textsubscript{2} mole fraction $\partial \chi_c/\partial t$ (\textmu mol mol\textsuperscript{-1} s\textsuperscript{-1}) as predicted by the linear or exponential fit (LI-COR, 2012).

### 6.3.4 Other environmental parameters

Each soil gas flux chamber was equipped with an in-built air temperature and atmospheric pressure sensor. A photosynthetically active radiation (PAR) sensor (LI-190R Quantum Sensor, LI-COR Biosciences, Inc.) was installed next to the control chamber. The data from these three sensors were recorded by the LI-COR system.

PAR readings were used as a proxy to distinguish day/night cycles. A tipping bucket rain gauge (Casella CEL, Inc.) was deployed in the vicinity of the soakaway to measure precipitation. During setup 1, additional soil sensors (GS3, Decagon Devices, Inc.) were inserted into the surface soil next to each chamber for continuous measurements of VWC, electrical conductivity (EC), and soil temperature with 5 min intervals. Bulk soil EC as recorded by the soil sensors was converted to pore water EC as described in METER (2018).
6.3.5 Data analysis

Data analysis was performed using R, version 3.4.3 (R Core Team, 2014). Significance levels were set to 0.05. Data were examined for evidence of normality and homogeneity and, if needed, log-transformed to attain normality. Post-hoc Tukey’s tests and confidence intervals were used to address the statistical significance of differences in observed CO$_2$ and CH$_4$ fluxes from the soakaway to fluxes observed over the control soil.

6.4 Results

6.4.1 Overall CO$_2$ and CH$_4$ fluxes

To assess the spatio-temporal distribution of both CO$_2$ and CH$_4$ fluxes over the septic tank soakaway, automated flux measurements were conducted on hourly intervals for a total of 81 days from a set of six distinct sampling locations distributed over the soakaway plus one control over natural soil.

Observed soakaway CO$_2$ fluxes (collars 1 to 6, see Figure 6.1) during this time ranged from 0.66 to 22.72 μmol CO$_2$ m$^{-2}$ s$^{-1}$ with a median of 7.28 μmol CO$_2$ m$^{-2}$ s$^{-1}$ (see Figure 6.2). Control fluxes (collar C, see Figure 6.1) observed during the same period exhibited lower overall median CO$_2$ fluxes of 6.40 μmol CO$_2$ m$^{-2}$ s$^{-1}$ and an overall slightly narrower range of observed fluxes from 1.19 to 19.92 μmol CO$_2$ m$^{-2}$ s$^{-1}$.

Observed CH$_4$ fluxes from the soakaway (collars 1 to 6, see Figure 6.1) during this time expressed a wide range from −6.72 to 553.33 nmol CH$_4$ m$^{-2}$ s$^{-1}$ with a median of −0.28 nmol CH$_4$ m$^{-2}$ s$^{-1}$, representing a median net take-up of CH$_4$ over the soakaway with occasional high emission events close to the inlet (see Figure 6.2). Control fluxes (collar C, see Figure 6.1) observed during the same period, exhibited lower overall median CH$_4$ fluxes of −0.67 nmol CH$_4$ m$^{-2}$ s$^{-1}$ representing a continuous uptake of CH$_4$ with fluxes ranging from −2.91 to −0.13 nmol CH$_4$ m$^{-2}$ s$^{-1}$.
6. CO$_2$ AND CH$_4$ EMISSIONS FROM SEPTIC TANK SOAKAWAY

Figure 6.2: Distribution of observed (A) carbon dioxide and (B) methane fluxes from hourly measurements over the soakaway and control soil for setup 1 and setup 2. Note, the x-axis for methane fluxes is cuberoot transformed for improved visualization.
Figure 6.3: Plan view of the spatial distribution of median (A) carbon dioxide and (B) methane fluxes over the soakaway (collars 1 to 6) and control soil. Effluent reaches the soakaway (gray area with adjacent zone of influence in light gray) in direction of the arrow from the septic tank (ST).
6.4.2 Spatial distribution of fluxes

To assess the spatial distribution of CO$_2$ and CH$_4$ fluxes over the soakaway, two different setups of chamber placements were deployed in succession. In setup 1 fluxes along the natural gradient were observed and in setup 2 two of the chambers were moved to capture the lateral variation of flux values.

In setup 1, the highest median CO$_2$ flux ($8.70 \mu$mol CO$_2$ m$^{-2}$ s$^{-1}$) was observed at collar 4, located 4 m downhill from the septic tank. The lowest median CO$_2$ flux ($5.54 \mu$mol CO$_2$ m$^{-2}$ s$^{-1}$) was observed at collar 3, located 3 m downhill from the septic tank outlet pipe, and was, interestingly, lower than the median flux from the control soil ($6.61 \mu$mol CO$_2$ m$^{-2}$ s$^{-1}$) recorded over the same period of time (see Figure 6.2). With the exception of the sampling point located closest to the septic tank, all CO$_2$ fluxes observed during the deployment of setup 1 were found to be significantly different than the control (post-hoc Tukey’s test, $p < 0.05$).

In setup 2, the highest median CO$_2$ flux ($9.38 \mu$mol CO$_2$ m$^{-2}$ s$^{-1}$) was again observed at collar 4 and the lowest median CO$_2$ flux ($5.24 \mu$mol CO$_2$ m$^{-2}$ s$^{-1}$) was observed at collar 1 located closest the effluent discharge point. Observed median control fluxes were similar to the ones observed during setup 1 ($6.23 \mu$mol CO$_2$ m$^{-2}$ s$^{-1}$). However, fluxes from collars 5 and 6, located 1 m away from collar 2 (towards the left of the expected effluent flow direction) and 3 (towards the right of the expected effluent flow direction), respectively, did not express significantly different fluxes compared to control soil (post-hoc Tukey’s test, $p < 0.05$).

In setup 1, the highest median CH$_4$ flux was observed at collar 1, located closest to the septic tank, with $6.49 \text{nmol CH}_4 \text{m}^{-2} \text{s}^{-1}$. Median CH$_4$ fluxes from the other chambers decreased continuously with distance from the septic tank with $0.58$, $-0.38$ and $-0.52 \text{nmol CH}_4 \text{m}^{-2} \text{s}^{-1}$ at location 2, 3 and 4, respectively, and resulted in an estimated net CH$_4$ uptake at the collars located further than 2 m away from the septic tank. Despite the overall median uptake at collar 3, however, occasional CH$_4$ emission events of up to $9.15 \text{nmol CH}_4 \text{m}^{-2} \text{s}^{-1}$ were recorded (see Figures 6.2 and 6.3). Collar 4 and the control soil, on the other hand, only expressed CH$_4$ uptake throughout all observations in setup 1, with median control CH$_4$ fluxes of $-0.73 \text{nmol CH}_4 \text{m}^{-2} \text{s}^{-1}$. Overall, only the fluxes recorded at the two collars located closest to the septic tank expressed net CH$_4$ emissions that were significantly different to the control (post-hoc Tukey’s test, $p < 0.05$).

In setup 2, CH$_4$ emissions events were observed only at the collar located closest to the septic tank with median fluxes of $18.67 \text{nmol CH}_4 \text{m}^{-2} \text{s}^{-1}$, exceeding those observed during setup 1 at the same location by nearly 3-fold. All other sampling locations expressed a
net CH$_4$ uptake throughout the deployment of setup 2 and were not significantly different to control fluxes.

### 6.4.3 Temporal variation of fluxes

Overall temporal variations of observed CO$_2$ fluxes over the soakaway generally followed patterns similar in trend and magnitude to the control, except for the last two weeks of setup 2 where soakaway CO$_2$ fluxes increased over control fluxes (see Figure 6.4). For most parts, soakaway CO$_2$ fluxes mimicked natural trends related to environmental parameters such as soil and air temperature rather than being driven by effluent discharge which acts as an effective addition of C to the soil biozone but also drives changes in soil VWC and pore water EC. Diurnal variations in both soakaway and control CO$_2$ fluxes were strongly correlated to soil temperature and, thus, microbial activity (see Table 6.3). Elevated CO$_2$ fluxes were recorded during day time from both the soakaway and control (see Figure 6.5), similar to previous research conducted on this site (Somlai-Haase et al., 2017). Median CO$_2$ fluxes started to increase from approximately 7:00 to 8:00 in the morning and peaked between 14:00 to 16:00 in the afternoon before receding to lower overall fluxes overnight.
Figure 6.4: Time series of observed (A) carbon dioxide and (B) methane fluxes over the soakaway (sampling point 4 as example for carbon dioxide and sampling point 1 as example for methane) and control soil for setup 1 and setup 2. Note the difference in scaling the y–axes for methane fluxes. Gray lines visualize long-term trends obtained by local regression of the individual data points.

CH$_4$ fluxes

Soakaway CH$_4$ fluxes, in general, were less correlated to environmentally driven changes than control CH$_4$ fluxes (see Table 6.4). Fluxes observed at collar 1, closest to the septic tank, were significantly higher than fluxes of any of the other sampling points over the soakaway and control soil (post-hoc Tukey’s test, $p < 0.05$), with emission rates at this location regularly (i.e. 97% of the time) exceeding those at other locations (see Figure 6.3). Diurnal soil gas flux patterns were less pronounced for CH$_4$ compared to CO$_2$ (see Figure 6.3). While the control soil exhibited its lowest median emissions between 7:00 and 8:00 in the morning and peak median emissions between 11:00 to 14:00, median CH$_4$ fluxes recorded at all other times of the day were not significantly different from each other (based on group–wise comparison of confidence intervals). Similar trends were not observed at any
Figure 6.5: Diurnal variations of observed fluxes over collar 4 and collar 1 for (A) carbon dioxide and (B) methane, respectively, as well as fluxes observed over control soil. Thin gray lines represent measurements from individual days. Thick gray lines represent hourly median fluxes. Collar 4 and collar 1 are chosen as example for soakaway fluxes as they represent the highest overall median fluxes for carbon dioxide and methane, respectively.

of the sampling points located over the soakaway. The lack of distinct high and low peaks in intra–day patterns coupled with significantly higher inter–day variability of observed median soakaway CH$_4$ fluxes indicates that non–environmental forces must have driven soakaway CH$_4$ fluxes. E.g., single periods of extremely high methane emissions (up to 60–times higher compared to median CH$_4$ fluxes) were recorded over collar 1, located closest to the septic tank, during a 3–day period between 21$^{st}$ and 24$^{th}$ April 2016 (see Figure 6.5). Almost 30% of the total CH$_4$ emissions from collar 1 recorded during this study were released during this short period. Soil sensor data and visual assessment during flux chamber installation indicated that, until the occurrence of this event the soil adjacent to the collar had been saturated and exhibited partially flooded conditions in the vicinity of the collar. However significant reductions of soil VWC and pore water EC were recorded
at the onset of the emission event. It is, thus, hypothesized that CH$_4$ was degassed as the upper soil layer dried out. After the degassing event, CH$_4$ emissions ceased as the soil profile continued to dry until partial rewetting of the soil occurred following a single precipitation event on 26$^{th}$ April, while CO$_2$ emissions remained unaffected.

![Graph showing methane (CH$_4$) and carbon dioxide (CO$_2$) fluxes under drying and typical conditions](image)

**Figure 6.6:** Extreme methane emission event during drying conditions following saturated soil conditions (left) and typical methane flux fluctuations during variably saturated conditions for comparison (right) from collar 1. While carbon dioxide fluxes appear to follow strong diurnal fluctuations driven mainly by soil temperature, methane emission events appear to be driven by a change in soil volumetric water content.

### 6.4.4 Environmental drivers of carbon dioxide and methane fluxes

Positive correlations were identified between CO$_2$ fluxes and soil temperature and negative correlations between CO$_2$ fluxes and soil VWC and pore water EC, explaining 43% to 67%, 14% to 32%, and 14% to 40% of changes in CO$_2$ flux, respectively, for both soakaway and control fluxes (see Table 6.3). While the influence of soil temperature on soakaway CO$_2$ fluxes within the first 2 m from the septic tank did not express significant differences compared to observed fluxes over control soil, control fluxes expressed stronger correlations to soil temperature than soakaway fluxes at these locations (with $R^2$ of 0.67, 0.59 and 0.53 for control, 1 m distance and 2 m distance, respectively). Correlations of CO$_2$ flux to soil
VWC were generally weak for both control and soakaway positions. Only collar 4, located farthest away from the septic tank, expressed significantly different CO$_2$ flux responses to changes in soil VWC compared to the control. Pore water EC had overall weak to moderate correlations to CO$_2$ fluxes with all collars located 2 m or further from the septic tank expressing significantly higher sensitivity to changes in pore water EC compared to the control.

As indicated by the fact that soakaway CH$_4$ fluxes expressed distinctively different temporal behavior to CO$_2$ and control fluxes (see Figures 6.4 and 6.5), considerably weaker correlations between observed CH$_4$ fluxes over the soakaway and environmental parameters were identified (see Table 6.3). While 36, 59 and 62% of changes in observed control CH$_4$ flux could be explained by changes in soil temperature, VWC and pore water EC, respectively, those parameters only accounted for a maximum of 3, 19 and 20% of changes in observed soakaway CH$_4$ fluxes. While CH$_4$ fluxes observed over all collars located 2 m or further from the septic tank expressed the same directional correlations for environmental parameters to observed CH$_4$ fluxes as the control soil (i.e., negative correlations for soil temperature and positive correlations for soil VWC and pore water EC; although with varying degrees of sensitivity), collar 1 expressed either no response (soil VWC and pore water EC) or an inverse response (soil temperature) to environmental inputs.

<table>
<thead>
<tr>
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<th>Distance</th>
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<th>Min</th>
<th>Max</th>
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</thead>
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<tr>
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<td>0.51</td>
</tr>
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<td>1.07</td>
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<td>1.96</td>
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Table 6.3: Correlations between observed carbon dioxide (CO₂) and methane (CH₄) fluxes and environmental parameters during setup 1 for sampling points over the soakaway located 1, 2, 3 and 4 m from the septic tank and control soil. Regression coefficients are given as point estimate of the linear slope ± 95% confidence interval. Coefficients at locations marked with † were significantly different (α = 0.05) from the control for the respective GHG and environmental parameter. Coefficients at locations marked with ‡ were not significantly different from zero, i.e. linear independence.

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<th>GHG Parameter</th>
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<th>R²</th>
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<td>0.59 ± 0.03</td>
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</table>
6.4. RESULTS

6.4.5 Spatio–temporal variability of observed fluxes

Daily totals of observed CO₂ fluxes expressed a wider spatio–temporal variability compared to CH₄ fluxes with CO₂ emissions generally increasing and CH₄ emissions generally decreasing with increasing distance from the septic tank (see Figure 6.7).

Daily total CO₂ emissions ranged from 8.37 to 54.29 g CO₂ m⁻² d⁻¹ with an overall median of 27.40 g CO₂ m⁻² d⁻¹. While collars 1, 2 and 4 expressed comparable CO₂ flux patterns, median fluxes from collar 3 were found to be lower than emissions from the control soil, emphasizing the high spatial variability of emissions detected over the soakaway. Elevated median CO₂ emissions were observed at collar 4, located farthest away from the septic tank, where aerobic conditions were more likely to occur temporarily due to the effluent plume not continuously reaching the upper soil layers – soil VWC data obtained from soil sensors installed adjacent to the collars recorded mean soil VWC values of 0.457, 0.436, 0.402 and 0.392 m³ m⁻³ for collars 1, 2, 3 and 4, respectively. However, occasional events of high positive CO₂ fluxes, i.e. emissions, were observed close to the septic tank. E.g., in early May 2016, elevated levels CO₂ emissions were recorded over all collars within the soakaway and receded in the following two weeks from up to 52.53 g CO₂ m⁻² d⁻¹ to a median of 32.02 g CO₂ m⁻² d⁻¹ within the first 2 m of the soakaway while remaining relatively constant with a median of 40.99 g CO₂ m⁻² d⁻¹ at a distance of 4 m from the septic tank for the same period of time (see Figure 6.7).

Daily total CH₄ fluxes expressed distinctly different flux patterns compared to CO₂ fluxes with generally lower temporal but more distinct spatial variability and ranged from −0.001 g CH₄ m⁻² d⁻¹ (i.e. net uptake) to 0.454 g CH₄ m⁻² d⁻¹ (i.e. net emissions) with an overall median of −0.0001 g CH₄ m⁻² d⁻¹ over the soakaway. However, as described earlier, short periods of high CH₄ emissions were observed over collar 1, located closest to the septic tank, and were linked to changes in soil VWC. Significant daily total CH₄ emissions were limited to collars located within the first 1 m from the septic tank throughout the study, with occasional daily total emissions recorded at 2 m distance.
6. CO₂ AND CH₄ EMISSIONS FROM SEPTIC TANK SOAKAWAY

Figure 6.7: Spatio-temporal distribution of daily total (A) carbon dioxide and (B) methane fluxes over the soakaway during setup 1. Values are linearly interpolated between observations (collar 1 – collar 4) in both temporal (x-axis as day of the year) and spatial (y-axis as distance from the septic tank) direction. Note, values on the colour scale for methane fluxes are cuberoot transformed for improved visualisation.

6.4.6 Discussions and Conclusions

Septic systems used for on-site wastewater treatment are potential sources of groundwater and atmospheric pollution. While the contaminant transport and attenuation in the receiving soil has been the focus of a wide range of studies over the past few decades, gaseous emissions from on-site installations in the field have attracted little targeted research so far. Additionally, studies on GHG emissions escaping to the atmosphere from septic system STUs were limited in their scope by either assuming a spatio-temporal (Leverenz et al., 2010; Diaz-Valbuena et al., 2011; Truhlar et al., 2016) or spatial (Somlai-Haase et al., 2017) homogeneity of flux distributions. The research presented in this study provides, to our knowledge, the first attempt to integrate continuously quantified emissions of both CO₂ and CH₄ from an STU (in the form of a soakaway) using hourly observations with spatially distributed measurements along a series of sections across the effective percolation area.

Automated long-term CO₂ and CH₄ soil gas flux measurements were conducted semi-continuously over a period of 81 days using two distinct spatially distributed chamber
6.4. RESULTS

Deployment setups: in setup 1 fluxes were recorded along the natural gradient of the site and expected dominant effluent dispersal direction, in setup 2 chambers were moved to capture the potential lateral spread of effluent in and GHG emissions from the soil.

Both CO₂ and CH₄ fluxes observed over the soakaway expressed distinct temporal variability. While CO₂ fluxes were mainly driven by diurnal changes in soil temperature, environmental parameters did not influence observed CH₄ fluxes, except for periods of substantial changes in VWC. Almost 30% of the total CH₄ emissions recorded during this study from the sampling location closest to the septic tank were released within a 3-day period at the onset of extended dry conditions with observed peak fluxes up to 60-times higher compared to the overall median. It is hypothesized that significant amounts of CH₄ that were trapped in the soil profile were degassed during this event as the upper soil layer dried out. As CH₄ gas bubbles get entrapped in the soil above and within the microbially active zone of the soakaway under saturated conditions, changes in hydraulic conductivity and effective pore structure can potentially alter effluent dispersal patterns and affect substrate distribution and pollutant attenuation in the soil profile. Similar CH₄ degassing events from variably saturated soils during drying events and changes of ambient pressure have been reported in peat soils and fens (Strack et al., 2005; Forbrich et al., 2010; Swenson et al., 2013), paddy fields (Denier van der Gon et al., 1996; Han et al., 2005) and landfill sites (Jones and Nedwell, 1993; McPhillips et al., 2016), however, found elevated CH₄ emissions in partially ponded roadside ditches dominantly after storm events, i.e. saturated conditions after wetting events. After the degassing event, CH₄ emissions ceased as the soil profile continued to dry until partial rewetting of the soil occurred following a single rainfall event several days later. Cai et al. (2016) suggested that high concentrations of CH₄ produced within anaerobic pockets under saturated conditions could slowly diffuse upward during drying conditions, inducing rapid growth of methanotrophs at the aerobic/anaerobic interface of the surface soil. As CH₄ production requires strictly anaerobic conditions, the extended dry conditions below a critical VWC level apparently resulted in sufficient availability of oxygen in the soil profile and potential inhibition of methanogenic activity. Well aerated soils are shown to be sinks for atmospheric CH₄ through microbial oxidation in the upper soil layer (Smith et al., 2018).

CO₂ fluxes were affected mostly by changes in soil temperature and followed distinct diurnal patterns with higher emissions occurring during day time. However, relatively high CO₂ emission events were observed occasionally throughout the soakaway with maximum emissions limited to approximately 2-fold increases over median levels. In a previous study though, Somlai-Haase et al. (2017) reported extended periods of elevated CO₂ fluxes up to 20-times higher than overall median fluxes from the same system at a single location over the soakaway. This high temporal variability emphasizes the importance of high-frequency,
long–term measurements of soil gas fluxes over STUs while including both environmental (such as meteorological and soil–physical data) and non-environmental (such as microbial community studies and effluent and substrate loadings) drivers into GHG flux models from STUs in order to make predictions about their responses to environmental changes.

Changes in the spatial distribution of both CO\textsubscript{2} and CH\textsubscript{4} fluxes were more pronounced along the natural gradient of the site and expected dominant effluent dispersal direction. Fluxes recorded perpendicular to the gradient expressed little to no differences compared to control soil. While median CO\textsubscript{2} fluxes expressed no clear pattern with distance from the septic tank, net CH\textsubscript{4} emissions were limited to the first 2 m. However, the effective lack of recorded CH\textsubscript{4} emissions over the soakaway at distances \(>\) 2 m from the septic tank may be due to the presence of CH\textsubscript{4} oxidation microbial communities in the upper soil layer as observed mean VWCs decreased with distance from the septic tank. Oxidation of methane by bacteria within the soil profile has been reported previously from both landfill sites (Jones and Nedwell, 1993) and STUs (Fernández-Baca et al., 2018).

In a previous study on CO\textsubscript{2} emissions from this site, Somlai-Haase et al. (2017) used the distribution of indicator vegetation on the soil surface and soil pore water quality data from Gill et al. (2013) as a proxy to estimate the effective soakaway area being approximately 6 m\textsuperscript{2}. However, the results from this study with spatially distributed soil gas flux measurements indicates that the microbiologically active zone in the soil may have temporarily extended further than the original soakaway area at depth, resulting in the highest CO\textsubscript{2} emissions measured 4 m downhill from the effluent discharge location. Assuming the original soakaway area and an average of two occupants in the household, total CO\textsubscript{2} emissions from the soakaway ranged from 7.52 g CO\textsubscript{2} cap\textsuperscript{-1} d\textsuperscript{-1} to 259.12 g CO\textsubscript{2} cap\textsuperscript{-1} d\textsuperscript{-1} with a median of 83.03 g CO\textsubscript{2} cap\textsuperscript{-1} d\textsuperscript{-1} and CH\textsubscript{4} emissions from the soakaway ranged from −0.03 g CH\textsubscript{4} cap\textsuperscript{-1} d\textsuperscript{-1} to 2.30 g CH\textsubscript{4} cap\textsuperscript{-1} d\textsuperscript{-1} with a median of −0.001 g CH\textsubscript{4} cap\textsuperscript{-1} d\textsuperscript{-1}. Both fluxes are comparable to fluxes presented by Truhlar et al. (2016) who reported CO\textsubscript{2} emissions ranging from −140 g CO\textsubscript{2} cap\textsuperscript{-1} d\textsuperscript{-1} to 450 g CO\textsubscript{2} cap\textsuperscript{-1} d\textsuperscript{-1} with a mean of 130 g CO\textsubscript{2} cap\textsuperscript{-1} d\textsuperscript{-1} and CH\textsubscript{4} emissions ranging from −0.11 g CH\textsubscript{4} cap\textsuperscript{-1} d\textsuperscript{-1} to 0.16 g CH\textsubscript{4} cap\textsuperscript{-1} d\textsuperscript{-1} with a mean of −0.0038 g CH\textsubscript{4} cap\textsuperscript{-1} d\textsuperscript{-1} from trench–based and sand filter STUs. Flux values reported by Truhlar et al. (2016) expressed a wider overall range for CO\textsubscript{2} fluxes, potentially resulting from the differences in temperature and VWC experienced between the respective sites. However, the maximum CH\textsubscript{4} fluxes were one order of magnitude lower than the ones observed in this study during the high emission events close to the septic tank. Assuming homogeneous spatial flux distributions, median CO\textsubscript{2} fluxes presented previously by Somlai-Haase et al. (2017) from collars positioned centrally over the soakaway were 78.22 g CO\textsubscript{2} cap\textsuperscript{-1} d\textsuperscript{-1}, i.e. 6.1% lower than median values observed in this study. While Truhlar et al. (2016) did not
specify the location of their sampling positions over the STUs, the spatial distribution of both CO\textsubscript{2} and CH\textsubscript{4} fluxes observed in this study empathizes the importance of deploying multi–chamber setups in order to more accurately capture localized shifts in effluent dispersal, VWC conditions and potential changes in microbially mediated C transformation pathways.

Despite occasional high emission events, the soakaway acted as a weak net consumer of CH\textsubscript{4} with an overall uptake similar to fluxes observed over urban lawns (Groffman and Pouyat, 2009; Livesey et al., 2010) and the majority of sites reported in Fernández-Baca et al. (2018) who found that in the upper layers of the soil methanotrophic populations were more abundant than methanogenic populations. However, the soakaway consumed overall 0.033 kg CO\textsubscript{2}Eq. yr\textsuperscript{−1} less CH\textsubscript{4} and emitted 7.327 kg CO\textsubscript{2} yr\textsuperscript{−1} more CO\textsubscript{2} than a similarly sized area of control soil which is approximately 2–times less than previously estimated (Somlai-Haase et al., 2017) and which demonstrates the importance of long–term spatio–temporal monitoring.

In summary, this study demonstrated that there are distinct spatio–temporal patterns for both CO\textsubscript{2} and CH\textsubscript{4} fluxes observed over a septic tank soakaway driven by both environmental factors and sub–surface effluent dispersal. In order to enhance our understanding of the environmental impacts of such engineered nature–based solutions future studies should deploy long–term, multi–chamber setups over a wider range of STU technology choices such as constructed wetlands, trench–based STUs or evaporation systems. To estimate total system emissions, integrated studies combining soil gas flux measurements over the STU with the quantification of GHG fluxes directly from the septic tank surface and vent system are needed to more accurately capture the spatial and temporal heterogeneity of gas fluxes from on–site installations.

6.5 Acknowledgements

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Author contributions

L.G. conceived the study and secured funding. C.S., J.K. and L.G. planned the experiments. C.S. and J.K. performed the experiments and analyzed data. C.S., J.K. and L.G. wrote the manuscript. All authors interpreted the results and reviewed the manuscript.
Greenhouse gas emissions from different stages and levels of on-site wastewater treatment systems

Celia Somlai, Jan Knappe and Laurence Gill

This paper is in preparation for submission.
Schematic drawing of the research sites with estimated total net GHG emissions from full wastewater treatment systems and emissions from different portion of on-site wastewater treatment systems with relevant contribution of the different gases (CO\(_2\) – blue, CH\(_4\) – red and N\(_2\)O – grey). Negative values indicate net reduction in emissions.

### 7.1 Abstract

In this study, a complex analysis of two domestic on-site wastewater treatment systems (DWWTSs) is presented in order to quantify their greenhouse gas (GHG) emissions. The different stages of the treatment process (i.e. septic tank and soil treatment unit), are compared including how the use of up-front packaged secondary treatment units impact on the net emissions from soil treatment units. GHG emissions were variable from the different parts of the DWWTSs and between the two different sites. The total net emissions from the full systems were 17.0 and 21.9 CO\(_{2}\)Eq. kg cap\(^{-1}\) yr\(^{-1}\) at the two different sites, respectively. Over 80% of the total net emissions was in the form of CO\(_2\), around 15% in CH\(_4\) and less than 2% in N\(_2\)O. At one of the sites where a rotating biofilter contactor was used as secondary treatment, a significant difference was observed between net emissions comparing septic tank effluent and secondary effluent. 39.6 CO\(_{2}\)Eq. kg cap\(^{-1}\) yr\(^{-1}\) total net emissions were estimated from the system receiving septic tank effluent and 5.6 CO\(_{2}\)Eq. kg cap\(^{-1}\) yr\(^{-1}\) net reduction from the system receiving secondary effluent compared with control soil. At the other site, no significant differences were observed between systems receiving septic tank or secondary effluent of coconut husk filter.
7.2 Introduction

Over the last few decades, a wide range of research studies on greenhouse gas (GHG) emissions have reported that wastewater treatment systems are potential sources of GHG emissions ([Metcalf and Tchobanoglous, 1972; Tchobanoglous et al., 1990; Czepiel et al., 1992, 1995; El-Fadel and Massoud, 2001; Tallec et al., 2006; Frijns et al., 2008; Hofman et al., 2011; Law et al., 2012; Daelman et al., 2013, 2015; Hwang et al., 2016; Pascale et al., 2017]). Overall GHG emissions from the waste and wastewater sector are estimated to be around 2% of the total estimated national emissions in the Republic of Ireland, ([EPA, 2018a]). Globally the wastewater sector is estimated to cause up to 1.5% of total GHG and 5% of non–CO\textsubscript{2} GHG emissions, and is expected to contribute 42% to overall waste-related GHG emissions by 2030, compared to 36% in 1990 ([Bogner et al., 2008; USEPA, 2012]). The quantification of direct GHG emissions from wastewater treatment systems is currently based on the application of estimation methodologies that have been published by the Intergovernmental Panel on Climate Change ([IPCC, 2013]). However, these estimations are considered highly uncertain as they are based on a limited number of case studies and rely heavily on secondary assumptions rather than primary data.

Most research in this area to date, however, has focused on large-scale, centralised treatment systems ([Czepiel et al., 1993, 1995; Johansson et al., 2004; Cakir and Stenstrom, 2005; Yoshida et al., 2014; Masuda et al., 2015; Kwok et al., 2015]). Despite an estimated 20% of the population relying on on-site wastewater treatment and disposal in both the European Economic Area and the United States ([EEA, 2013; USEPA, 2016]), only a limited number of studies, so far, have been based on direct measurements of GHG emissions from on-site and decentralized wastewater treatment systems ([Leverenz et al., 2010; Diaz-Valbuena et al., 2011; Truhlár et al., 2016; Somlai-Haase et al., 2017; Somlai et al., 2019]).

In low- and middle-income countries, the share of these systems is significantly higher than the global average and estimated to provide improved sanitation for up to 64% of the population ([Hawkins et al., 2014]). With an ever-growing global population, predominantly in areas currently unserved by centralised sanitation solutions, there is a general imperative to build and improve sanitation infrastructure globally as manifested in the Sustainable Development Goal (SDG) 6 on water and sanitation. Septic systems, commonly consisting of a septic tank (ST) for initial collection, storage and partial treatment of domestic wastewater followed usually by some form of soil treatment unit (STU) for additional treatment and disposal into subsurface soil, are environmentally and economically sustainable on-site solutions for wastewater treatment, if properly constructed and managed. Recently in Ireland, there is a shift towards implementing an increasing number of packaged treatment systems over traditional septic systems ([CSO, 2017]). These packaged
systems require to carry out operation and maintenance of the system in accordance with the manufacturers instructions including regular servicing, desludging, and replacement of consumables such as filter media (EPA, 2009). One typical type of packaged treatments systems based on a fixed biofilm process, in which the effluent is put into contact with a microbial biofilm growing naturally, for example rotating biological contactor or media filter.

As septic systems have simple design and rely on naturally occurring biogeochemical processes for wastewater treatment, they are inherently suitable for off-the-grid solutions and in areas with disperse settlement patterns, however; these biogeochemical processes are rather complex and involve a wide range of microbial populations and dissolved O₂ concentrations (Beal et al., 2005; Tomaras et al., 2009). Wilhelm et al. (1994) presented a conceptual model describing two main redox environments for on-site systems. The first redox zone – ST – is characterised as an anaerobic environment with initially high concentrations of organic C and N, and low dissolved O₂ levels, where the organic matter is mainly degraded via hydrolysis, acidogenesis, and methanogenesis producing both CO₂ and CH₄. In the second redox zone – STU – both aerobic and anaerobic conditions can exist, depending on the availability of O₂. Gaseous diffusion in the unsaturated zone receiving the partially treated effluent supplies O₂ for aerobic oxidation of organic C resulting in the production of CO₂. In STU trenches, a biomat forms at the infiltrative surface with time and gradually clogs soil pores. The development of this biomat layer is linked to the physical accumulation of suspended solids within soil pores and microbial growth (Jones and Taylor, 1965; Thomas et al., 1966; Bouma, 1975; Siegrist and Boyle, 1987; Beach et al., 2005; McKinley and Siegrist, 2010). As gradual biomat development results in a growing resistance to flow and reductions in hydraulic conductivity, ponding of effluent at the trench base can occur and anaerobic conditions may develop (Siegrist and Boyle, 1987; Siegrist et al., 2000; Van Cuyk et al., 2001; Beach et al., 2005; Hu et al., 2007). In the absence of O₂, e.g. in saturated microsites, ponded infiltration trenches, or within mature biomats at the infiltrative surface, anaerobic organisms such as methanogenic bacteria break down insoluble organic compounds to CO₂ and CH₄. Research by Fernández-Baca et al. (2018) indicated recently that elevated concentrations of CH₄ in the upper soil layer above the infiltration trenches, can sustain a population of CH₄ consuming bacteria, which may act to reduce overall net CH₄ fluxes from the soil to the atmosphere.

Only a few studies have investigated the GHG emissions from septic systems (Diaz-Valbuena et al., 2011; Truhlar et al., 2016; Somlai-Haase et al., 2017; Somlai et al., 2019). Diaz-Valbuena et al. (2011) focused on the STs of eight conventional septic systems and based on the limited number of measurements over the two STUs, reported 33.3 g CO₂ cap⁻¹ d⁻¹, 11.0 g CH₄ cap⁻¹ d⁻¹ and 0.005 g N₂O cap⁻¹ d⁻¹ emissions from the
STs and negligible emissions from the STUs. [Truhlar et al. (2016)] investigated the sand filters, STUs and vent outlets of eight septic systems and found mean CH$_4$ uptake ($-0.004 \text{ g CH}_4 \text{ cap}^{-1} \text{ d}^{-1}$) and mean CO$_2$ and N$_2$O emissions (130 g CO$_2$ cap$^{-1}$ d$^{-1}$ and 0.022 g N$_2$O cap$^{-1}$ d$^{-1}$) from STUs. [Somlai-Haase et al. (2017)] deployed a single soil CO$_2$ flux chamber for a period of 17 months over a ST soakaway to quantify long-term and diurnal CO$_2$ flux variations and found fluxes ranging from 0.43$\mu$mol CO$_2$ m$^{-2}$ s$^{-1}$ to 100.26$\mu$mol CO$_2$ m$^{-2}$ s$^{-1}$ with a median of 6.86$\mu$mol CO$_2$ m$^{-2}$ s$^{-1}$. [Somlai et al. (2019)] measured CO$_2$ and CH$_4$ fluxes over the same ST soakaway using a multi-chamber automated soil gas flux chamber system with high spatial and temporal resolution. The soakaway was found to be a net emitter of both CO$_2$ and CH$_4$, releasing a total of 7.327 kg CO$_2$ yr$^{-1}$ and 0.033 kg CO$_2$Eq. yr$^{-1}$, respectively. However, localised CH$_4$ degassing events were observed during drying conditions with up to 60 times higher fluxes compared to the overall median.

The main aim of this paper to compare emission rates of different levels of on-site wastewater treatment at two different sites; from septic tanks as well as from the STU receiving septic tank effluent and secondary treated effluent, from two fixed film packaged treatment plants.

7.3 Materials and methods

7.3.1 Study sites and treatment systems

Two research sites were constructed serving single detached houses in Co. Limerick, the Republic of Ireland: A gravity–flow based on-site treatment system was installed in Crecora (CC) and a pumped–flow system was installed in Kilmallock (KM) due to the lack of a natural gradient from the house in KM towards the STU area designated for final effluent discharge to soil, see Figure [Diagram]. All sites were instrumented with a network of soil sensors, pore water samplers and soil collars for gas flux measurements.
A two-compartment, pre-fabricated septic tank (Aswasep NS4S, Molloy Precast Ltd., Ireland) was installed as primary treatment module for both sites. The primary effluent (PE) from the septic tank was then split equally with one half directly fed into the first two trenches of a soil treatment unit (STU) and the other half fed into a packaged secondary treatment system before being discharged as secondary effluent (SE) into the other two STU trenches. Both secondary units were based on attached growth biofilm processes. While in CC a rotating biological reactor, RBC, (Klargester BioDisc, Kingspan Ltd., UK) was used, the pumped flow regime in KM allowed for the deployment of a coconut husk peat media filter (Ecoflo, Premier Tech Aqua, France). See Table 7.1 for details on both study sites and wastewater treatment systems. By splitting the effluent equally between primary and secondary fed STU trenches allows for the direct comparison of the influence of pre-treatment on system performance and microbially mediated GHG emissions with identical subsoil characteristics on the respective research sites.

### 7.3.2 GHG flux measurements

**Equipment and flux calculations**

Long-term soil gas flux measurements of CO₂ were carried out using an automated system consisting of cylindrical opaque long-term chambers (LI8100-104, LI-COR Biosciences,
Table 7.1: Site and wastewater treatment system characterisation.

<table>
<thead>
<tr>
<th>Site</th>
<th>KM</th>
<th>CC</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aquifer category</td>
<td>LI(^a)</td>
<td>Rkd(^b)</td>
</tr>
<tr>
<td>Groundwater vulnerability</td>
<td>Medium</td>
<td>High</td>
</tr>
<tr>
<td>Topsoil</td>
<td>BminDW(^c)</td>
<td>BminDW(^c)</td>
</tr>
<tr>
<td>Subsoil</td>
<td>Clay loam</td>
<td>Sandy loam</td>
</tr>
<tr>
<td>Area usage</td>
<td>Lawn</td>
<td>Pasture land</td>
</tr>
<tr>
<td>Climate</td>
<td>maritime Cfb(^d)</td>
<td>maritime Cfb(^d)</td>
</tr>
<tr>
<td>Average annual temperature(^e)</td>
<td>10 °C</td>
<td>10 °C</td>
</tr>
<tr>
<td>Average annual rainfall(^e)</td>
<td>1200 mm</td>
<td>1200 mm</td>
</tr>
<tr>
<td>Total annual sunshine(^e)</td>
<td>1250 h</td>
<td>1250 h</td>
</tr>
</tbody>
</table>

System

<table>
<thead>
<tr>
<th></th>
<th>KM</th>
<th>CC</th>
</tr>
</thead>
<tbody>
<tr>
<td>No. of occupants</td>
<td>4</td>
<td>4</td>
</tr>
<tr>
<td>ST</td>
<td>Two-chamber</td>
<td>Two-chamber</td>
</tr>
<tr>
<td>Capacity (m(^3))</td>
<td>6.2</td>
<td>6.2</td>
</tr>
<tr>
<td>Desludged</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>HRT(^f)(d)</td>
<td>10.3</td>
<td>10.3</td>
</tr>
<tr>
<td>Secondary treatment</td>
<td>Coconut filter</td>
<td>RBC</td>
</tr>
<tr>
<td>Effluent</td>
<td>Pumped</td>
<td>Gravity flow</td>
</tr>
<tr>
<td>STU</td>
<td>2 trenches - PE</td>
<td>2 trenches - PE</td>
</tr>
<tr>
<td>Construction</td>
<td>2015</td>
<td>2016</td>
</tr>
<tr>
<td>STU Area (m(^2))</td>
<td>180</td>
<td>180</td>
</tr>
<tr>
<td>(K_{fs}) (m d(^{-1}))</td>
<td>0.32</td>
<td>0.18</td>
</tr>
</tbody>
</table>

\(^a\) Deep well drained mineral soil derived from mainly basic;  
\(^b\) Regionally Important Aquifer - Karstified (diffuse);  
\(^c\) Deep well drained mineral soil derived from mainly basic;  
\(^d\) Warm temperate, fully humid, warm summer \(\text{(Kottek et al., 2006)}\);  
\(^e\) \(\text{(Walsh, 2012)}\);  
\(^f\) Hydraulic Retention Time;
Inc.), a multiplexer, and a non-dispersive infrared gas analyser for analysis of CO$_2$ (LI-8100A Automated Soil Gas Flux System, LI-COR Biosciences, Inc.). A gas analyser capable of CH$_4$ detection (Ultraportable Greenhouse Gas Analyser, model 915-0011, Los Gatos Research) was integrated into the gas loop according to instructions provided by Li-Cor (2015). A detailed description of the long-term soil gas flux system employed in this study can be found in Somlar-Haase et al. (2017). Additionally, regular gas flux measurements from the liquid surface of both chambers of the septic tank and STU vent system were conducted using a survey chamber and vent pipe cap connected to the gas sampling loop, respectively.

Both linear and exponential models were fit to the gas concentration data using the Soil-FluxPro 4.0 software package (LI-COR Biosciences, Inc.). Regression $R^2$-values were used as quality control and selection criterion for using either the linear or exponential model for flux calculations (the model with higher $R^2$-value was selected). Observations resulting in model fits with low overall $R^2$-values ($R^2 < 0.9$ for CO$_2$ and $R^2 < 0.8$ for CH$_4$) were assumed compromised and discarded. It has been suggested that flux values estimated by linear models tend to systematically underestimate actual fluxes while exponential models based on diffusion theory result in more accurate flux calculations (Healy et al., 1996; Nakano et al., 2004; Levy et al., 2011). Exponential models, though, are not able to capture net uptake phenomena (e.g. for CH$_4$) and are generally less robust compared to linear models. However, Forbrich et al. (2010) observed that while for CH$_4$ non-linear changes of concentration over time occurred mainly during periods of changing VWC, overall, linear models were the best-fitted model for a majority of CH$_4$ measurements across sites with a variety of soil VWCs.

Gas fluxes $F$ were calculated from the initial change in concentration for each incubation using a mass balance approach

$$F = \frac{V_{sys} p_0 (1 - \chi_w)}{R s T_0} \cdot \frac{\partial \chi_c(t)}{\partial t}$$

with the total system volume $V_{sys}$ (0.06 m$^3$), the atmospheric pressure at the beginning of the measurement $p_0$ (Pa), the chamber air water vapor mole fraction $\chi_w$ (mol mol$^{-1}$), the universal gas constant $R$ (8.314 Pa m$^3$ K$^{-1}$ mol$^{-1}$), the soil collar surface area $s$ (0.032 m$^2$), the absolute temperature at the beginning of the measurement $T_0$ (K), and the initial change of chamber water vapor corrected CO$_2$ mole fraction $\partial \chi_c/\partial t$ (µmol mol$^{-1}$ s$^{-1}$) as predicted by the linear or exponential fit (LI-COR, 2012).
The emission rates were calculated from the change in concentration passing through the analysers over time and normalised to areal units (Grinham et al., 2011).

Measurements of dissolved gas concentrations in the STs and gas-phase concentrations within the passive vent systems at the ends of each plastic distribution pipe were also directly sampled. Additional gas samples were collected and transported back to the laboratory for further analyses of CH$_4$ and N$_2$O. All fluxes were scaled using either the area of the water surface in the ST, STU, or pipe vents, as appropriate, and converted into a mass flux of gas per capita (in g cap$^{-1}$ d$^{-1}$) using the ideal gas law and information about the number of residents in each household, see SI [D.1] for details.

**Septic tank**

In the ST, two different setups were used to measure CO$_2$ and CH$_4$ gas fluxes; (i) discrete survey measurements carried out manually whilst on site and (ii) diurnal measurements carried out by automated survey flux chamber. Discrete survey measurements for CO$_2$ were carried out 13 times in CC and 14 times in KM between May 2017 and July 2018. Due to the availability of the CH$_4$ analyser, discrete flux measurements of CH$_4$ were limited to 8 times at CC and 9 times at KM. Diurnal measurements were carried out in the second chamber of the ST in CC between 20 and 21 November 2017 and in KM between 24 and 25 July 2018 with 5-minute frequency.

In order to measure GHG fluxes from the water surface of ST, a sampling setup was adopted from Leverenz et al. (2011), where a collar was placed in to the septic tank to hold the chamber and create a gas loop between the water surface and gas analyser during measurements. This collar was composed of a rigid PVC pipe (inner diameter 20.3 cm, outer diameter 21.3 cm O.D. and length 19 cm) and supported by three legs (length 210 cm) going to the bottom of the tank. The collars were placed such that they extended from beneath to above the water level of the ST. These inserts were left in place for the duration of the experiment to prevent disturbance in the septic tank.

During flux incubations, a survey chamber (LI-8100-103 20 cm Survey Chamber, LI-COR Biosciences, Inc.), that was connected to an analyser control unit with two sets of LI-COR extensions (length: 15 m), was lowered down to the collar in the ST. The lids of the ST was opened in each compartment. One set of the extensions connected the gas lines, the other set of extensions was controlling the survey chamber and providing the gas channel for lowering down and lifting up the survey chamber over the collar between measurements. A pressure/vacuum air flow system expands and contracts a bellows to raise and lower the chambers over a collar to conduct the flux measurement.
Additional to the flux measurements, water samples were collected to estimate the dissolved CO$_2$ and CH$_4$ concentrations and water temperature, EC, pH and dissolved O$_2$ were measured. Dissolved CO$_2$ and CH$_4$ concentrations were estimated using the headspace method ([Hope et al., 1995]) and the UGGA Ultraportable Greenhouse Gas Analyser. 100 ml water samples were collected in a 250 ml glass bottle, the bottle was sealed with the cup and inlet and outlet pipe were attached to the cap. The pipes were directly connected to the UGGA, the analyser bubbled out the dissolved gases to enable the equilibrium concentration of the gases in the bottle system to be measured. The temperature, EC, pH and dissolved O$_2$ were measured using a WTW multi-parameter portable meter (WTW ProfiLine Multi 3320, WTW GmbG, Germany).

**Soil treatment unit**

Two different methods were used to measure CO$_2$ and CH$_4$ soil fluxes from the STU; (i) discrete survey measurements carried out manually whilst on site and (ii) diurnal measurements carried out by automated long-term flux chambers. Additional to the flux measurements, gas samples were collected and carried to the laboratory to measure CH$_4$ and N$_2$O flux with a gas chromatography.

In total 24 collars were distributed over the STU and control soil at each site; 5 collars per trench and additionally 4 collars were set up outside of the STU over control soil, see Figure C.2 in Appendix. The collars were installed in the ground, leaving ca. 0.05 m above ground on which flux chambers were placed creating a seal between the collar and the chamber.

The discrete flux measurements of CO$_2$ with the survey chamber (LI-8100-103, LI-COR Biosciences, Inc.) were carried out at 15 occasions at CC and at 13 occasions at KM between May 2017 and July 2018. Due to the availability of the CH$_4$ analyser, discrete flux measurements of CH$_4$ were limited to 8 times at CC and 5 times at KM. Due to time constriction of manual handling of samples being analysed with gas chromatograph, a limited number of samples were taken for measuring N$_2$O fluxes. Between July 2017 and July 2018 on 8 occasions at both sites, one collar per trench was sampled.

For the diurnal measurements, 5 cylindrical opaque long-term chambers (LI8100-104, LI-COR Biosciences, Inc.) were deployed; 1 chamber over each trench and 1 over the control soil rotating between the sites and between different collars. The flux system consisted of three main components: gas analysers, multiplexer (LI-8150-8) and multiple long-term chambers (LI-8100-104 and LI-8100-104C). The basic concept of the measurement is that chamber is moved by a non-flexible arm over a collar which is inserted into the soil creating
a gas loop between the chamber and the analyser to be able to measure the change in gas concentrations under the chamber. The detailed description of long-term soil flux system can be found in (Somlai-Haase et al., 2017).

Additional gas samples were collected from the long term chambers and taken back to the laboratory for further gas chromatography analysis of CH$_4$ and N$_2$O. , see below.

**Vent system**

Effluent distribution pipes embedded in the four gravel trenches were connected to individual above-surface vent systems by elbows fitted at the end of each pipe. Gases escaping through these vent were captured using a Vent Wizard 800+ (in-house development) consisting of a sealed cap with a single gas line that can be connected to the gas sampling loop. For each measurement, the protective end vent terminal was replaced with the Vent Wizard 800+ and the system was incubated for at least 3 min and until no further increase in gas concentration was observed. Immediately before each incubation, a hotwire anemometer (LU8050, TQC Sheen, The Netherlands) was inserted into the vent pipe to measure average temperature inside and undisturbed air velocity from the vent as the mean of 30 measurements with 2s interval to eliminate potential disturbances by wind gusts. Gas fluxes from the vent were determined by multiplying the final steady-state gas concentration observed inside the vent with the surface area of the vent port and measured air velocity. A total of 8 and 9 CO$_2$ flux measurements were conducted per vent in CC and KM, respectively. CH$_4$ fluxes were determined in the field from 3 and 4 measurements per vent in CC and KM, respectively, due to limited instrument availability. Additional gas samples were collected at 6 occasions from each site and taken to the Environmental Engineering Lab at Trinity College Dublin for further analysis of CH$_4$ and N$_2$O.

**Gas samples for CH$_4$ and N$_2$O**

Additional gas samples were collected from the air stream of the flux measurements integrating a Tee-fitting with a septum (8100-664 Trace Gas Sample Kit) in the gas loop. Four consecutive gas samples were collected during a period of 6-9 minutes using a 50 ml plastic syringe with a needle to withdraw gas and injecting into sealed and pre-evacuated gas vials (20 ml mL GC vials). The gas samples were analysed within maximum 72 hours using a Perkin Elmer Clarus 500 gas chromatograph (Perkin Elmer, Waltham, MA, USA) equipped with capillary columns (Elite-Plot Q), flame ionisation detector (FID) and electron capture detectors (ECD).
7.3.3 Auxilliary data

Meteorological data were recorded on all sites using a full weather station (Campbell Scientific Ltd, UK) capturing mean air temperature, barometric pressure, net radiation, rainfall, relative humidity, wind direction, and wind speed at 2 m height.

7.3.4 Data collection and processing

Weather station, tipping bucket and soil sensor data were collected with hourly intervals using a CR1000 data logger (Campbell Scientific Ltd, UK). Meteorological data was used to calculate standardized reference evapotranspiration ET\textsubscript{0} based on energy balance and mass transfer principles using the ASCE Penman-Monteith (ASCE-PM) method for hourly timesteps and short crop reference (Allen et al., 1998; Shahidian et al., 2012). All data analysis was performed using R, version 3.4.3 (R Core Team, 2014).

7.4 Results and Discussion

7.4.1 Effluent quantity and quality

A mean daily wastewater production of 498.8 l d\textsuperscript{-1} (i.e., 124.7 l cap\textsuperscript{-1} d\textsuperscript{-1}) and 288.2 l d\textsuperscript{-1} (i.e., 72.1 l cap\textsuperscript{-1} d\textsuperscript{-1}) was recorded in CC and KM, respectively, with overall uniform daily effluent production, except for distinct events following pump failure (in KM) and holidays (CC and KM) that equate to durations of < 3 weeks in total per site during this study.

7.4.2 GHG fluxes from septic tank

At both sites, the mean CO\textsubscript{2} fluxes in the first chamber of the ST (in CC 6.61 and in KM 3.69 μmol CO\textsubscript{2} m\textsuperscript{-2} s\textsuperscript{-1}) exceeded the fluxes in the second chamber (in CC 3.80 and in KM 2.39 μmol CO\textsubscript{2} m\textsuperscript{-2} s\textsuperscript{-1}) with wider range of fluxes in the first chamber (in CC 1.18 - 18.52 and in KM 1.22 - 8.05 μmol CO\textsubscript{2} m\textsuperscript{-2} s\textsuperscript{-1}) than in the second chamber (in CC 1.15 - 7.85 and in KM 1.02 - 4.81 μmol CO\textsubscript{2} m\textsuperscript{-2} s\textsuperscript{-1}), as shown in Figure 7.2a. The dissolved mean CO\textsubscript{2} concentrations were also higher in the first chamber in CC (24.23 mg CO\textsubscript{2} l\textsuperscript{-1}) than in the second chambers (21.81 mg CO\textsubscript{2} l\textsuperscript{-1}). However, in KM the mean CO\textsubscript{2} concentrations were lower in the first chamber (16.05 mg CO\textsubscript{2} l\textsuperscript{-1}) than in the second chambers (18.79 mg CO\textsubscript{2} l\textsuperscript{-1}).
The mean CH$_4$ fluxes in the first chamber were higher in CC (0.48 μmol CH$_4$ m$^{-2}$ s$^{-1}$) and lower in KM (0.43 μmol CH$_4$ m$^{-2}$ s$^{-1}$) than in the second chamber (in CC 0.31 and in KM 0.48 μmol CH$_4$ m$^{-2}$ s$^{-1}$), see Figure 7.2c. The mean dissolved CH$_4$ concentrations exceeded in the second chambers in both sites (mean 408.79 in CC and 491.95 μg CH$_4$ l$^{-1}$ in KM) exceeded the concentrations in the first chambers (mean 387.96 in CC and 213.56 μg CH$_4$ l$^{-1}$ in KM), see Figure 7.2d. It should be noted that a scum layer developed only at CC in first chamber.

However, these figures may be an underestimation of emissions from the ST. During a diurnal deployment of the survey chamber in the second chamber of the ST at KM (between 2018-07-24 19:00 and 2018-07-25 19:00), median of the CH$_4$ fluxes were almost 3 times higher than the median of the discrete measurements from the same chamber and the maximum CH$_4$ flux during this deployment exceeded more than 4 times the maximum discrete CH$_4$ flux, see Figure D.2f in Appendix. These high flux values during the diurnal deployment can be the result of capturing ebullition of CH$_4$, which has been found to be episodic in nature (Bastviken et al., 2011; Natchimuthu, 2016).
The difference between the CO$_2$ and CH$_4$ fluxes and dissolved concentrations might be explained with the two different stages of the anaerobic oxidation combined with the different water usage in the two different households and scum development. The anaerobic oxidation, where in the first stage the organic matters are converted to simple organic compounds and volatile fatty acids producing mostly CO$_2$ and in the second stage the soluble organic acids (which will have been leached out into the bulk liquid and could have passed into the second chamber) are stabilised by methanogens and most of the CH$_4$ is produced. Another research study into sludge accumulation in septic tanks carried out at the same sites (Gill et al., 2018), showed that, the first chambers at both STs had over 25 times higher sludge mass accumulation. However, the family living in KM used the washing machine at least once a day which could result the dilution of the organic concentration of their wastewater, this might be resulted the CO$_2$ flux rates in CC in comparison to KM are most likely due to the 170% higher carbon load of the wastewater.
in CC. In the ST in KM, there was no scum development observed, in contrary; in CC there was a really thick scum layer (up to 35 cm) developed in the first chamber by the end of first year after construction. The median dissolved O$_2$ were consistent in all the chambers on both sites (0.2 mg l$^{-1}$).

### 7.4.3 GHG fluxes from soil treatment unit

#### Discrete measurements

In CC, the mean (range) CO$_2$ fluxes were 4.03 (0.27 - 18.36), 3.01 (0.30 - 15.62) and 4.42 (0.45 - 15.56) μmol CO$_2$ m$^{-2}$ s$^{-1}$ from the two trenches receiving PE, the two trenches receiving SE and control area, respectively. In KM, the mean (range) CO$_2$ fluxes were 3.16 (0.13 - 11.78), 3.46 (0.12 - 18.15) and 2.73 (0.35 - 8.03) μmol CO$_2$ m$^{-2}$ s$^{-1}$ from the two trenches receiving PE, the two trenches receiving SE and control area, respectively. Interestingly, the highest mean CO$_2$ fluxes occurred in CC from the control area exceeding the ones from the STU and the lowest mean in KM from the control area, see Figure 7.3a. CO$_2$ fluxes from the STU are not significantly different from each other and they are within the range of fluxes from the control sites. This matches what was found by Diaz-Valbuena et al. (2011) who found negligible CO$_2$ emissions from the STUs they studied.

In CC, the mean (range) CH$_4$ fluxes were 11.60 (-8.01 - 852.86), −0.12 (-3.31 - 4.44) and −0.35 (-3.36 - 0) nmol CH$_4$ m$^{-2}$ s$^{-1}$ from the trenches receiving PE, the trenches receiving SE and control area, respectively. In KM, the mean (range) CH$_4$ fluxes were 21.59 (-2.67 - 703.57), 12.53 (-0.70 - 437.40) and −0.28 (-2.46 - 0) nmol CH$_4$ m$^{-2}$ s$^{-1}$ from the trenches receiving PE, the trenches receiving SE and control area, respectively. CH$_4$ fluxes from the control areas at both sites were continuously negative. This means that the control soils consumed CH$_4$ which is a similar finding to other studies on soils of grasslands and unfertilised pastures reported by Mosier et al. (1991, 1997); Dunfield et al. (1995); Braun et al. (2013). The highest median CH$_4$ fluxes occurred in KM at trench 1 and trench 3, see Figure 7.3b.

In CC, the mean (range) N$_2$O fluxes were 0.003 (-0.13 - 0.10), 0.016 (-0.16 - 0.15) and 0.048 (-0.02 - 0.27) nmol N$_2$O m$^{-2}$ s$^{-1}$ from the trenches receiving PE, the trenches receiving SE and control area, respectively. In KM, the mean (range) N$_2$O fluxes were 0.021 (-0.38 - 0.29), 0.005 (-0.63 - 0.35) and −0.073 (-0.38 -0.03) nmol N$_2$O m$^{-2}$ s$^{-1}$ from the trenches receiving PE, the trenches receiving SE and control area, respectively, see Figure 7.3c. The highest mean N$_2$O fluxes were measured in CC from the control area, in contrary, mean N$_2$O uptake was observed in KM from the control area. This can result from the
heterogeneity of the soil and the different soil types at the two different sites. As presence of plants affect on C and N dynamics in soil, and on the microbial processes that transform C and N. For example, plants add C to the soil via roots, roots exudates, and leaves, all of which can increase CO$_2$ flux. Processing of this material consumes O$_2$, which may result in hypoxia or anoxia in some areas of the soil, affecting N$_2$O and CH$_4$ dynamics. Roots also respire, producing CO$_2$ and some plants may emit N$_2$O and CH$_4$ naturally (Chen et al., 1999; Pihlatie et al., 2005; Bowatte et al., 2014). Bowatte et al. (2014) has shown that pasture plants can act as plant-mediated emitters of N$_2$O largely through acting as a conduit for emissions generated in the soil, which are also controlled by plants. Ammonia-oxidising bacteria have been found on the leaves of plants, on Norway spruce (Henriksen et al., 1999).
et al., 2007), on weeds in rice paddies (Bowatte et al., 2006) and on the pasture grass Lolium perenne (Bowatte et al., 2013). As the control soil and the STU and was used as grazing pasture in CC prior to the study, this could result the difference between the two sites. Note that studies focus on N₂O emissions have longer incubation period of 30 minute instead 6 – 9 minutes.

Our measurements demonstrate that CH₄ emissions from STUs can be up to 1000 folds higher than emissions from lawns and forested park areas, however; the N₂O emissions are in a narrower range than found in other studies. In order to compare these results to other studies, the fluxes values were scaled (for CO₂ µmol CO₂ m⁻² s⁻¹, for CH₄ nmol CH₄ m⁻² s⁻¹ and for N₂O nmol N₂O m⁻² s⁻¹, see in Table 7.2. A constructed STU consists of a series of perforated pipes laid in parallel trenches (approximately 45 cm) and backfilled with gravel (EPA, 2013) compare to a soakaway is a large pit back filled with stone and rubble that are receiving effluent from the ST by a single gravity fed effluent distribution point (EPA, 2013). Of the two gravity flow dispersal mechanisms, constructed STUs disperse effluent more broadly and at a shallower depth. This allows for better effluent treatment and decreases risk of groundwater contamination, making leach fields the preferred OWTS effluent dispersal technology. In STUs, wastewater is perlocated over a greater area than in soakaways, which means that there is an increased opportunity for effluent interaction with soil microbes, and the greater availability of oxygenated environments. This can be the result of the different of GHG emissions from STUs than from soakaways.

**Spatial variations of GHG fluxes**

Visualisation of the spatial distribution of mean CO₂ fluxes in CC shows that the collars placed over the trenches receiving PE had higher flux values than ones over the trenches receiving SE, see Figure 7.4a. In KM, clear difference in fluxes was not observed from trenches receiving PE and SE effluent, see Figure 7.4c. From the control area, really high mean CO₂ fluxes were measured, in KM the CO₂ fluxes from the control area were much lower.

The spatial distribution of mean CH₄ fluxes had no clear trend, few hotspots can be identify in Figure 7.4b and 7.4d.
Table 7.2: Summary of reported GHG flux ranges from soil treatment units and grass lawns.

<table>
<thead>
<tr>
<th></th>
<th>CO₂</th>
<th>CH₄</th>
<th>N₂O</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>μmol CO₂ m⁻² s⁻¹</td>
<td>nmol CH₄ m⁻² s⁻¹</td>
<td>nmol N₂O m⁻² s⁻¹</td>
</tr>
<tr>
<td><strong>Previous studies</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>STU⁵</td>
<td>negligible</td>
<td>negligible</td>
<td>negligible</td>
</tr>
<tr>
<td>STU, filter and control lawn⁶</td>
<td>-4.63 – 6.94</td>
<td>-9.26 – 2.32</td>
<td>-0.01 – 7.94</td>
</tr>
<tr>
<td>soakaway⁷</td>
<td>0.66 – 22.72</td>
<td>-6.72 – 553.33</td>
<td>-</td>
</tr>
<tr>
<td>control lawn ⁸</td>
<td>1.19 – 19.92</td>
<td>-2.91 – 0.13</td>
<td>-</td>
</tr>
<tr>
<td>grass lawn ⁹</td>
<td>-</td>
<td>-0.12 – 0.06</td>
<td>0.00 – 0.30</td>
</tr>
<tr>
<td>grass and forest plots</td>
<td>0.00 – 5.79e</td>
<td>-2.32 – 0.93e</td>
<td>-0.40 – 1.79f</td>
</tr>
<tr>
<td><strong>This study</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CC STU PE</td>
<td>0.27 – 18.36</td>
<td>-8.01 – 852.86</td>
<td>-0.13 – 0.10</td>
</tr>
<tr>
<td>CC STU SE</td>
<td>0.30 – 15.62</td>
<td>-3.31 – 4.44</td>
<td>-0.16 – 0.15</td>
</tr>
<tr>
<td>CC control grass lawn</td>
<td>0.45 – 15.56</td>
<td>-3.36 – 0</td>
<td>-0.02 – 0.27</td>
</tr>
<tr>
<td>KM STU PE</td>
<td>0.13 – 11.78</td>
<td>-2.67 – 703.57</td>
<td>-0.38 – 0.29</td>
</tr>
<tr>
<td>KM STU SE</td>
<td>0.12 – 18.15</td>
<td>-0.70 – 437.40</td>
<td>-0.63 – 0.35</td>
</tr>
<tr>
<td>KM control grass lawn</td>
<td>0.35 – 8.03</td>
<td>-2.46 – 0</td>
<td>-0.38 – 0.03</td>
</tr>
</tbody>
</table>

⁵ Diaz-Vallejo et al. (2013); ⁶ Trihlar et al. (2016); ⁷ Somlai et al. (2019); ⁸ Livesley et al. (2010); ⁹ Groffman et al. (2009); ¹⁰ Groffman and Pouyat (2009).
Figure 7.4: Spatial distribution of mean fluxes of CO₂ and CH₄ showing the locations of the measurements.
Diurnal variations of GHG fluxes

The diurnal variations of the GHG fluxes were investigated by calculating the hourly means for each long-term flux measurement campaign and then aggregating the fluxes by the trenches, see Figure 7.2 and 7.3. For CO$_2$ fluxes, a clear temperature dependence is observed, lower fluxes with narrower ranges during the colder months (from September until February) and higher fluxes with wider ranges over the warmer months. During the summer months (June, July and August), the fluxes from the control area exceeded the fluxes from the STU.

For CH$_4$ fluxes, no clear pattern was observed. However, higher hourly CH$_4$ fluxes were measured from trenches receiving PE.

7.4.4 GHG fluxes from passive vent systems

The overall mean of CO$_2$ fluxes from the vent systems in CC was 4 times higher than the mean of CO$_2$ in KM, 16.7 and 4.15 μmol CO$_2$ s$^{-1}$, respectively. The CO$_2$ fluxes had a much wider range at CC (0.87 - 170.15 μmol CO$_2$ s$^{-1}$) than in KM (0.12 - 20.07 μmol CO$_2$ s$^{-1}$), see Figure 7.5a. At both sites, means of CO$_2$ fluxes from the PE (CC: 26.64 and KM: 5.19 μmol CO$_2$ s$^{-1}$) exceeded the fluxes from the SE (CC: 6.74 and KM: 3.11 μmol CO$_2$ s$^{-1}$).

The overall mean of CH$_4$ fluxes from the vent systems in CC was over 10 times higher than those in KM; 43.6 and 3.66 nmol CH$_4$ s$^{-1}$, respectively, and CH$_4$ fluxes had a much wider range at CC (-0.80 - 318.00 nmol CH$_4$ s$^{-1}$) than in KM (-0.6 - 42 nmol CH$_4$ s$^{-1}$), see Figure 7.5b. At both field sites, means of CH$_4$ fluxes from the PE (CC: 82.9 and KM: 6.26 nmol CH$_4$ s$^{-1}$) highly exceeded the fluxes from the SE (CC: 0.01 and KM: -0.09 nmol CH$_4$ s$^{-1}$). However, the fluxes from the vent outlets receiving the same treated effluent were different, for example at CC site the mean CH$_4$ fluxes from trench 1 and 2 were 8.2 and 157.6 nmol CH$_4$ s$^{-1}$, respectively. CH$_4$ fluxes were constantly high from trench 2 at CC, similar to CO$_2$.

The overall means of N$_2$O fluxes from the vent systems in CC was over three times higher than those in KM, 1.26 and 0.37 nmol N$_2$O s$^{-1}$ at CC and KM, respectively. The N$_2$O fluxes had a much wider range at CC (-0.50 - 5.07 nmol N$_2$O s$^{-1}$) than in KM (0.04 - 1.25 nmol N$_2$O s$^{-1}$), see Figure 7.5c. At both sites, means of N$_2$O fluxes from the PE (CC: 1.65 and KM: 0.44 nmol N$_2$O s$^{-1}$) were higher than the fluxes from the SE (CC: 0.07 and KM: 0.13 nmol N$_2$O s$^{-1}$). N$_2$O fluxes were constantly high from trench 2 at CC highly exceeding the fluxes from the other vent outlets.
7.4. RESULTS AND DISCUSSION

Figure 7.5: (A) CO$_2$ flux, (B) CH$_4$ and (C) N$_2$O fluxes form the vent system outlets.

The significant differences between the flux rates from the vent system at the two sites can be due to the different organic load and different subsoil types. In CC, the mean organic load was much higher than in KM, resulting higher mean flux rates from the vent system in CC. In KM, the subsoil is more freely percolating soil than in CC, resulting in less retention and faster percolation through the subsoil to deeper layers.

In CC, trench 2 was a continuous GHG emitter with uniquely high values this can be linked to that the effluent spread much further in the trenches receiving PE, and it is nearer to the vent pipe. The reason for that can be the biomat spread further in the that trench. This one was proved also during a deconstructive soil sampling campaign.
in August 2018 (data from that field campaign is not discussed in this PhD research) by discovering really dark biomat formation at approximately 15 m further down along the trench, see Figure D.6 in Appendix.

7.4.5 Net GHG Emissions

Emissions from ST

The total estimated annual CO$_2$ emissions from the ST were 5.7 and 3.3 kg CO$_2$ cap$^{-1}$ yr$^{-1}$ at CC and KM, respectively, with higher CO$_2$ emissions in the first chamber at both sites. The total estimated annual CH$_4$ emissions from the ST were 0.11 and 0.12 kg CH$_4$ cap$^{-1}$ yr$^{-1}$ at CC and KM, respectively, with higher CH$_4$ emissions in the first chamber in CC and with higher CH$_4$ emissions in the second chamber in KM. Hence, the total annual CO$_2$ equivalent emissions from the STs were 8.4 and 6.3 kg CO$_2$Eq. cap$^{-1}$ yr$^{-1}$ at CC and KM, which are very similar. As it was stated above, these emissions from the ST may be an underestimation.

Net Emissions from STU

The total annual CO$_2$ emissions were estimated for the first 6 m (24 m$^2$) of the STUs where the collars were distributed using the mean fluxes for trenches receiving the same effluent for both site. The total annual CO$_2$ emissions from the trenches were 58.7 and 55.1 kg CO$_2$ cap$^{-1}$ yr$^{-1}$ at CC and KM, respectively, with slightly higher emissions from trenches receiving PE in CC and with slightly higher emissions from trenches receiving SE in KM. This could be due to the different packaged secondary treatment units. We did not measure CO$_2$ emission from the secondary treatment plants, which presumably will be high due to the amount of organics they break down. further the coconut husk filter provides an extra source of C for CO$_2$ emissions.

The net annual CO$_2$ emissions were calculated from the difference between the total annual CO$_2$ emissions and emissions from a similarly sized control area. Due to the high CO$_2$ fluxes from the control area in CC, the net annual CO$_2$ emissions were negative resulting a net redaction of 14.9 kg CO$_2$ cap$^{-1}$ yr$^{-1}$. In KM, the net annual CO$_2$ emissions were 9.6 kg CO$_2$ cap$^{-1}$ yr$^{-1}$.

The total annual CH$_4$ emissions from the STUs were 34.8 and 103.6 g CH$_4$ cap$^{-1}$ yr$^{-1}$ in CC and KM, respectively, with CH$_4$ uptake from trenches receiving SE in CC and high CH$_4$ production at trenches receiving PE in KM. The net annual CH$_4$ emissions from the
STUs were slightly higher (37.0 and 105.3 g CH\textsubscript{4} cap\textsuperscript{-1} yr\textsuperscript{-1}) than the total annual CH\textsubscript{4} emissions.

The total annual N\textsubscript{2}O emissions from the trenches were 0.16 and 0.22 g N\textsubscript{2}O cap\textsuperscript{-1} yr\textsuperscript{-1} at CC and KM, respectively, with significantly higher N\textsubscript{2}O emissions from trenches receiving SE in CC with significantly higher N\textsubscript{2}O emissions from trenches receiving PE in KM. Due to the high emissions from the control area, similar to CO\textsubscript{2} emissions, there was a net reduction in N\textsubscript{2}O emissions. The net annual N\textsubscript{2}O reduction was estimated to be 0.64 g N\textsubscript{2}O cap\textsuperscript{-1} yr\textsuperscript{-1} in CC in contrast with the 1.4 g N\textsubscript{2}O cap\textsuperscript{-1} yr\textsuperscript{-1} annual net emissions in KM.

Converting the net CH\textsubscript{4} and N\textsubscript{2}O emissions into kg CO\textsubscript{2}Eq. cap\textsuperscript{-1} yr\textsuperscript{-1} and summing up all the three gases, there was a net annual reduction of emissions of 14.2 kg CO\textsubscript{2}Eq. cap\textsuperscript{-1} yr\textsuperscript{-1} in CC and a net annual emissions of 12.7 kg CO\textsubscript{2}Eq. cap\textsuperscript{-1} yr\textsuperscript{-1} in KM.

**Emissions from vent system**

The total annual CO\textsubscript{2} emissions from the vent systems were 23.2 and 5.9 kg CO\textsubscript{2} cap\textsuperscript{-1} yr\textsuperscript{-1} at CC and KM, respectively. No clear pattern between CO\textsubscript{2} emissions from vent systems receiving primary or secondary effluent. The total gross annual CH\textsubscript{4} emissions from the vent systems were 20.97 and 1.6 g CH\textsubscript{4} cap\textsuperscript{-1} yr\textsuperscript{-1} at CC and KM, respectively, with significantly higher CH\textsubscript{4} emissions from trenches receiving primarily treated wastewater. The total gross annual N\textsubscript{2}O emissions from the vent systems were 1.2 and 0.4 g N\textsubscript{2}O cap\textsuperscript{-1} yr\textsuperscript{-1} at CC and KM, respectively, with higher N\textsubscript{2}O emissions from trenches receiving primarily treated wastewater. The total annual CO\textsubscript{2} equivalent emissions were 24.1 and 5.9 kg CO\textsubscript{2}Eq. cap\textsuperscript{-1} yr\textsuperscript{-1}, over 99% of the total emissions was in the form of CO\textsubscript{2}.

**Emissions from full septic systems**

The total net emissions from the full systems were 17.0 and 22.0 CO\textsubscript{2}Eq. kg cap\textsuperscript{-1} yr\textsuperscript{-1} in CC and KM, respectively. In CC, 81.8, 16.1 and 2.1% of the total net emissions were in the form of CO\textsubscript{2}, CH\textsubscript{4} and N\textsubscript{2}O as CO\textsubscript{2}Eq., In KM, the magnitude of the contribution of the different gases to the total net emissions was similar with 85.2, 14.3 and 0.5% of the total net emissions were CO\textsubscript{2}, CH\textsubscript{4} and N\textsubscript{2}O.

The research sites were set-up in order to be able to compare how inclusion of up-front packaged secondary treatment units impact on the net emissions from STUs. The net emissions were calculated by assuming that all four trenches receiving either only PE or
only SE from the packaged secondary treatment units. In CC, the difference between emissions from full systems receiving PE and SE was significant. The total net emissions were 39.6 CO$_2$Eq kg cap$^{-1}$ yr$^{-1}$ by assuming all four trenches receiving PE and the total net reduction in emissions yield 5.6 kg CO$_2$Eq cap$^{-1}$ yr$^{-1}$ uptake from the full system assuming all four trenches receiving SE. In KM, there was no significant difference between the two total net emissions, 21.0 and 22.9 kg CO$_2$Eq cap$^{-1}$ yr$^{-1}$.

It is important to note here that emission rates were not measured from the packaged secondary treatment units and they could not be included in this calculation. Also important to note that this study had a limitation using a single control chamber.

### 7.5 Conclusion

This research demonstrates that GHG emissions from the different parts of the DWWTSs are variable. The highest measured CO$_2$ flux rates were observed from the STUs at both sites, however; these rates were adjusted to account for the background soil emissions to calculate the net emissions from the STUs. Including the background soil concentrations into the estimates, the highest CO$_2$ emissions remained to be from the STU in KM and CO$_2$ reduction in emissions was observed with the highest CO$_2$ emissions from the vent system in CC. The STs contributed to the highest CH$_4$ emissions at both sites. In KM, high CH$_4$ emissions were observed also from the STU compared to the STU in CC. The highest N$_2$O production occurred in the vent systems at both sites. Vegetation was assumed to be homogeneous over the STU and control area, however; vegetation can be different in control area versus the STU and that can also influence the emission rates and spatial variations of the emissions. CH$_4$ production are well known to be promoted by waterlogged soils [Le Mer and Roger (2001)]. There is evidence that CH$_4$ consumption is more limited by CH$_4$ diffusion, and therefore by water-filled pore space, than by soil temperature [Ambus and Christensen, 1995], however; higher CH$_4$ emissions and uptake rates have been found by [Le Mer and Roger (2011)] with increasing temperature.

Other studies found positive correlations between N$_2$O emissions and increasing temperature. This is caused by a combined effect of the increased rate of enzymatic processes involved in nitrification and denitrification and increased rate of N$_2$O diffusion through the soil during the highest temperature periods. [Ryden et al., 1978; Ambus and Christensen, 1993; Butterbach-Bahl et al., 2013; Shurpali et al., 2016; Ambus and Christensen (1993); Smith et al. (1998)] found positive correlations between N$_2$O emissions and soil moisture as well.
8

CO₂, CH₄ and N₂O emissions from a willow evapotranspiration system treating domestic wastewater

Celia Somlai, Jan Knappe and Laurence Gill

This manuscript is in preparation for submission.

8.1 Abstract

In the Republic of Ireland, around 40% of the area is inadequate for percolation due to low permeability soil and subsoil, high water tables and/or low permeability bedrock. One solution to the treatment and disposal of on-site wastewater in such situations is the use of closed basin willow evapotranspiration (ET) systems in order to mitigate groundwater and surface water pollution in rural areas. In this study, greenhouse gas (CO₂, CH₄ and N₂O) fluxes were measured from one such willow ET system over four different quarters of the year between October 2017 and August 2018. Strong spatial and seasonal variations of the fluxes were observed. CO₂ and N₂O fluxes had similar seasonality, higher fluxes during early (1.09 – 5.18μmol CO₂ m⁻² s⁻¹ and -337.70 – 310 nmol N₂O m⁻² s⁻¹) and peak of growing seasons (1.14 – 48.26μmol CO₂ m⁻² s⁻¹ and 148.51 – 74.30 nmol N₂O m⁻² s⁻¹) and lower fluxes during end of the growing season (0.21 – 0.73μmol CO₂ m⁻² s⁻¹ and -93.07 – 74.30 nmol N₂O m⁻² s⁻¹) and during the dormant season (0.19 – 0.70μmol CO₂ m⁻² s⁻¹ and -246.72 – 61.70 nmol N₂O m⁻² s⁻¹). In contrary, the highest CH₄ fluxes were observed during end (-3.74 – 51.35 nmol CH₄ m⁻² s⁻¹) of the
8. GHG EMISSIONS FROM WILLOW SYSTEM

growing season (8.41 - 68.72 nmol CH₄ m⁻² s⁻¹). The total gross GHG emission from the evapotranspiration system (945 CO₂Eq. kg yr⁻¹ cap⁻¹) exceeded measured emissions from other on-site septic systems.

8.2 Introduction

In the Republic of Ireland, around 500,000 dwellings rely on on-site treatment and disposal of their wastewater, 87% of which use septic tanks followed by some form of percolation system (CSO, 2011). However, it is estimated that around 40% of the area of Ireland is inadequate for percolation due to low-permeability subsoils and bedrock and or insufficient attenuation due to high water tables and shallow subsoils (EPA, 2013). When permeability is insufficient to take the effluent load, ponding and breakout of untreated or partially treated effluent at the surface may occur, that might cause health risks as well as a risk of pollution to nearby surface waters. A potential solution for the treatment of on-site effluent in such regions of low subsoil permeability is to use evapotranspiration (ET) systems with willows trees. These systems consist of a septic tank and followed by sealed basins which are backfilled with soil into which willow trees are planted, see Figure 8.1. The concept is that the willow has a very high evapotranspiration rate up to 930 mm yr⁻¹ and the basins are designed to provide a sufficient ET capacity to minimise any net surface discharge from combined wastewater and rainfall loading over the course of a year. Such systems have been used in Denmark (Gregersen and Brix, 2001) and more recently trialled in Irish conditions (Curneen and Gill, 2014, 2015, 2016).

Willows are highly suitable for such an application in such temperate climates due to their high transpiration rates throughout the growing season (Rosenqvist et al., 1997; Pauliukonis and Schneider, 2001) efficient uptake of nutrients (Elowson, 1999; Dimitriou and Aronsson, 2011), tolerance of flooded soils and oxygen shortage in the root zone (Kuzovkina et al., 2003), and resilience to pollutants (Bialowiec et al., 2007).

In the recent trials in the Republic of Ireland, no system managed to achieve zero discharge in any year remaining at maximum level for much of the winter months, indicating some loss of water by lateral exfiltration at the surface (Curneen and Gill, 2014). However, chemical and microbiological sampling of the water in the sumps and ponded water over the winter periods showed good water quality, equivalent to surface runoff from the adjacent field, and so the systems were acting as excellent pollutant attenuation devices, even if they could not be described as fully zero discharge systems on an annual basis. Hence, in such Irish climatic conditions with high relative humidity and where low permeability
8.2. INTRODUCTION

subsoil is used to backfill the systems, it is not expected that this system will be zero discharge, but will discharge the rainfall runoff at times over the winter period (December to March).

Domestic wastewater is known to be a significant source of GHGs, however; there have been no studies on ET systems with regards to their greenhouse gas (GHG) emissions. In these systems, the wastewater leaves the household and enters first a septic tank, where organic matter ferments under anaerobic conditions, producing CO$_2$ and CH$_4$. Further in the septic tank, amino acids release nitrogen as ammonium (NH$_4^+$). The partially treated sewage from the septic tank flow to the ET basins where the environment is mixed aerobic and anaerobic depending on the water level and oxygen level in the basin. When there is sufficient oxygen and the water level is lower, NH$_4^+$ and organic molecules are oxidised, producing nitrate (NO$_3^-$) and CO$_2$, respectively. When the basin is saturated, biological denitrification occurs, reducing NO$_3^-$ to N$_2$. Incomplete nitrification and denitrification both contribute to N$_2$O emissions (Wilhelm et al., 1994; Richard et al., 2014).

![Figure 8.1: Schematic diagram of a closed basin willow evapotranspiration system receiving effluent from a septic tank.](image)

In this study, CO$_2$, CH$_4$ and N$_2$O fluxes were measured from a full-scale, closed-basin, willow ET system treating domestic wastewater over four different quarters of the year between October 2017 and August 2018 representing different periods of the willows' annual growth cycle.
8.3 Materials and methods

8.3.1 Study site

The study site is located in the North-East of the Republic of Ireland. The characterisation of the site is see Table 8.1. The top (above 25 cm) and subsoil (below 25 cm) are classified as AminPD (mineral poorly drained - mainly acid) and IrSTLPSsS (Irish Sea till derived from lower palaeozoic sandstones and shales), respectively. The classification of the aquifer is poor and groundwater vulnerability is low.

The climate is classified as maritime CFb (warm temperate, fully humid, warm summer) after Köppen-Geiger (Kottek et al., 2006). The mean annual temperature is 10°C with mean seasonal minimum and maximum between 4°C and 15°C in winter and summer, respectively; average annual rainfall is 700 mm without significant seasonal variation and 1250 h total annual sunshine (Walsh, 2012).

8.3.2 Wastewater Treatment System

The wastewater treatment system is used by a single household with 4 full-time inhabitants (i.e. 4 PE) and it is located the back garden of the family house, see Figure 8.2. The ET system was designed on the basis of extensive field trials on 13 full scale sealed willow systems by the Environmental Engineering Research Group in Trinity College Dublin (Curneen and Gill, 2015, 2016). These trials have been used to draft national guidelines for the Environmental Protection Agency. The design was based upon 1001cap⁻¹ d⁻¹ wastewater production, which is considerably lower than the figure of 1501cap⁻¹ d⁻¹ assumed in the Code of Practice (EPA, 2009). On this basis, the house was fitted with water saving devices, such as dual flush toilets, low flow shower heads and tap aerators to achieve the lower wastewater production. The wastewater treatment system was constructed in 2016.

The wastewater was collected, stored and pre-treated in a septic tank (2 chambers, capacity of 3.6 m³ and hydraulic retention time of 10.7 d). It should be noted that trials carried out by Curneen and Gill (2015) have demonstrated no advantage of discharging secondary effluent onto the willows compared to primary treated effluent; in fact willows dosed with primary treated effluent showed a sight enhancement on evapotranspiration rates due to the higher organics and nutrients. The effluent from the septic tank was gravity fed to a distribution box, where the effluent was equally divided between ET basins in parallel.
Table 8.1: Site and wastewater treatment system characterisation.

<table>
<thead>
<tr>
<th>Site characteristics</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Aquifer category</td>
<td>Pu&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td>Groundwater vulnerability</td>
<td>Low</td>
</tr>
<tr>
<td>Topsoil</td>
<td>AminPD&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>Subsoil</td>
<td>IrSTLPSs&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td>Area usage</td>
<td>Garden</td>
</tr>
<tr>
<td>Climate</td>
<td>Cfb&lt;sup&gt;d&lt;/sup&gt;</td>
</tr>
<tr>
<td>Average annual temperature</td>
<td>10 °C</td>
</tr>
<tr>
<td>Average annual rainfall</td>
<td>1200 mm</td>
</tr>
<tr>
<td>Total annual sunshine</td>
<td>1250 h</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>System characteristics</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>No. of occupants</td>
<td>4</td>
</tr>
<tr>
<td>Septic tank</td>
<td>Two-chamber</td>
</tr>
<tr>
<td>Capacity (m&lt;sup&gt;3&lt;/sup&gt;)</td>
<td>3.1</td>
</tr>
<tr>
<td>Desludged</td>
<td>-</td>
</tr>
<tr>
<td>HRT&lt;sup&gt;f&lt;/sup&gt;(d)</td>
<td>10.7</td>
</tr>
<tr>
<td>Effluent</td>
<td>Gravity flow</td>
</tr>
<tr>
<td>Soil treatment unit</td>
<td>Willow</td>
</tr>
<tr>
<td>Construction</td>
<td>2016</td>
</tr>
<tr>
<td>Area (m&lt;sup&gt;2&lt;/sup&gt;)</td>
<td>2 x 247</td>
</tr>
<tr>
<td>$K_{f_s}$ (m d&lt;sup&gt;-1&lt;/sup&gt;)</td>
<td>0.08</td>
</tr>
</tbody>
</table>

<sup>a</sup> Poor Aquifer - Bedrock which is Generally Unproductive;  
<sup>b</sup> Mineral poorly drained - Mainly acid;  
<sup>c</sup> Irish Sea Till derived from Lower Palaeozoic sandstones and shales;  
<sup>d</sup> Warm temperate, fully humid, warm summer;  
<sup>f</sup> Limestone till;  
<sup>f</sup> Hydraulic Retention Time;
The two basins (29 m long, 8.5 m wide and 1.8 m deep) were excavated next to each other. The basins were lined with an impermeable membrane and geosynthetic barriers on the both sides of the impermeable membrane. The wastewater was equally distributed into the two willow beds via two rows of 110 mm diameter rigid plastic pipes laid horizontally in a gravel layer (0.3 m at the bottom of the basin at a 3 m spacing).

The system incorporated an inspection well in a small excavated depression at the base of the system, in which the water level and water temperature were monitored using an OTT Orpheus water level logger. Each system also had an overflow pipe from the surface of the bed which discharged via a tipping bucket flow gauge used to quantity surface discharge. A bund was constructed along the perimeter of the willow system surface to create extra storage in case of excess surface flooding. The original soil was then backfilled into the basin, first the subsoil followed by topsoil at the end. Planting of willow cuttings was carried out in March 2016 at a density of 3 m$^{-2}$ using three different cultivars of willow (Salix viminalis). Measurements were conducted over one of the ET systems.

Water samples were taken from the septic tank effluent and the sumps inside the ET systems during each of the GHG sampling periods during each annual Quarter (see below).
8.3.3 Soil flux measurements

The GHG fluxes were directly measured from the soil surface of the willow bed by carrying out soil gas flux measurements of CO$_2$ using an automated system (LI-8100A Automated Soil Gas Flux System, LI-COR Biosciences, Inc.) consisting of a non-dispersive infrared gas analyser, a multiplexer, and 5 cylindrical opaque long-term chambers (LIS100-104, LI-COR Biosciences, Inc.) and depending on the availability with an additional gas analyser (UGGA Ultraportable Greenhouse Gas Analyser, model 915-0011, manufactured by Los Gatos Research) for CH$_4$. The detailed description of long-term soil flux system can be found in (Somlai-Haase et al., 2017) and in a paper in preparation, see Chapter 7. In order to measure the GHG emissions from the willow bed, different measurement setups were used. In total, 12 plastic collars (diameter 20 cm) were placed into the soil over the willow bed at 2, 7, 15 and 25 m from the effluent at the right, middle and left side and additional 3 collars were placed outside of the bed for control measurements, see Figure 8.3.

Three different methods were used to measure CO$_2$, CH$_4$ and N$_2$O gas fluxes; (i) discrete survey measurements carried out manually whilst on site, (ii) long-term diurnal measurements carried out by automated long-term flux chambers and (iii) gas samples collected on site and brought back to the laboratory for analysis by a gas chromatography for CH$_4$ and N$_2$O. The measurements were focussed into four intensive monitoring periods (quarters) to represent different periods of the willows’ annual growth cycle as follows:

- Q1 (autumn 2017 end of growing season): 22-25 October 2017
8. GHG EMISSIONS FROM WILLOW SYSTEM

- Q2 (early spring 2018 end of dormant season): 23-26 March 2018
- Q3 (early summer 2018 early growing season): 1-4 June 2018
- Q4 (mid summer 2018 peak growing season): 2-5 August 2018

Discrete flux measurements To measure the on-site survey CO₂ fluxes, an automated soil flux system (LI-8100A, LI-COR Biosciences, Inc.) consisting of a non-dispersive infrared gas analyser and a survey chamber (LI-8100-103, LI-COR Biosciences, Inc.) was used eight times during the four different quarters Q1 (19th October and 31st of October 2017), Q2 (22nd and 26th of March 2018), Q3 (31st May and 6th June 2018) and Q4 (30th July and 7th August 2018). On four occasions in the different quarters Q1 (19th October and 31st of October 2017) and Q4 (30th July and 7th August 2018), the CO₂ soil flux system was extended with an additional gas analyser (Ultraportable Greenhouse Gas Analyser, model 915-0011, manufactured by Los Gatos Research) in order to measure CH₄ fluxes as well.

Diurnal flux measurements To investigate the diurnal variation of CO₂ fluxes, five automated long-term flux chambers (LI-8100-104, LI-COR Biosciences, Inc.) were deployed four times during Q1, Q2, Q3 and Q4 for 3-day periods (72 hours from 8 am until 8 am) to perform hourly flux measurements. During Q1 and Q4, the additional CH₄ analyser was also deployed.

Discrete samples for laboratory GC analysis

During the four different quarters on specific days (19th October 2017, 22nd March, 6th June and 30th July 2017), gas samples were also collected and taken back to the laboratory for analysis by gas chromatography for CH₄ and N₂O.

8.3.4 Analysis

The flux values measured by the gas analysers were calculated using SoilFluxPro 4.0 software (Li-Cor Biosciences, Inc.). The gas chromatograph results were processed in Excel. Both calculations were based on a mass balance approach, detailed description given in Section D.1 in Appendix. R²-value of the regressions were used as quality control and either the linear or the exponential fit was used depending on the higher R²-value (CO₂: R² < 0.9, CH₄: R² < 0.8 for data recorded by the gas analysers and CH₄: R² < 0.7, N₂O:
R² < 0.7 for the manually collected samples and analysed by the gas chromatography). When both CO₂ analysers were deployed the means of the two measurements were used as well as when CH₄ analyser and gas samples were taken simultaneously, the average was used.

### 8.3.5 Environmental parameters

Meteorological data were recorded at hourly intervals of atmospheric pressure, air temperature, relative humidity, net radiation, wind speed, wind direction and precipitation (Campbell Scientific with CR1000 data logger). ET is calculated using ASCE Penman-Monteith on hourly time steps and short crop reference. Wind gust speed is maximum wind speed averaged over three seconds per hour. In order to measure the effluent flow, tipping bucket and reed switch (Casella CEL, Inc. UK) for gravity flow in the distribution box was used combined with a level logger for pump sumps, see Figure 8.4. During long-term flux measurements, soil temperature and VWC was also measured at each collar using 8100-202 Soil Moisture Probe and 8100-201 Soil Temperature Probe (LI-COR Biosciences, Inc.).

### 8.3.6 Water quality parameters

The septic tank effluent and willow system samples (taken from inspection well sumps within the system) were periodically analysed for chemical oxygen demand (COD), ammonium (NH₄⁺N), nitrite (NO₂⁻N), nitrate (NO₃⁻N), orthophosphate (PO₄³⁻P), chloride (Cl) and sulphate (SO₄²⁻) using a Merck Spectroquant Nova 60 spectrophotometer. In addition, total organic carbon (TOC) and total nitrogen (TN), were measured with a Shimadzu TOC-V CSN analyser (Shimadzu Scientific Instruments Inc., USA). Indicator bacteria of faecal contamination, Total Coliforms (TC) and E. coli were analysed for using the IDEXX Colilert-18 test with enumeration carried out using IDEXX Quanti-Tray/2000, a semi-automated quantification method based on the Standard Methods Most Probably Number (MPN) model.

### 8.3.7 Statistical analysis

All analysis was performed on R version 3.2.3 (R Core Team, 2013) with a significance level defined to be 0.05. All variables were examined for evidence of normality and homogeneity.
Post-hoc Tukey’s tests were used to address the significance of whether observed CO$_2$ and CH$_4$ fluxes were different from the fluxes observed at the control site.

GIS Inverse Distance Weighted (IDW) interpolation of a point vector layer (measurements from the collars) were used in QGIS (version 3.0.0-Girona), where the sample points are weighted during interpolation such that the influence of one point relative to another declines with distance from the unknown point you want to create. The distance coefficient power was set for 2, and the number of columns = 58 and number of rows = 17 resulting 0.5 m x 0.5 m grid cells.

8.4 Results and Discussion

8.4.1 Meteorological results and ET system performance

The mean air temperature was 10.1°C ranging between -3.0 and 27.2°C. The mean wind speed was 1.7 m s$^{-1}$ with the maximum value of 9.7 m s$^{-1}$. The mean relative humidity was 87.9% ranging between 33.7 to 100.0%. The mean water temperature was 9.8°C ranging between 6.1 and 13.7°C. The ET system behaved as expected with the water level in the sump starting to drop in spring and reaching minimum level (0.83 m) in mid summer. The total rainfall was 613 mm between October 2017 and August 2018 with hourly average precipitation of 0.1 mm h$^{-1}$, see Figure 8.4.
8.4. RESULTS AND DISCUSSION

Figure 8.4: Top, daily sum precipitation mm d$^{-1}$. Bottom, daily average water level (black line) and temperature (grey) in the inspection well with the black straight horizontal line indicating the soil surface in the willow bed. The horizontal lines shows the soil flux measurements (black dashed line - survey measurements and gas sampling, black continuous line - only survey measurements, grey lines - hourly long-term measurements).

8.4.2 Water quality

High concentrations of organic and nitrogenous materials entered the ET systems across the monitoring periods, as shown in Table 8.2. It appears that a significant fraction of these compounds are being removed by the time they reach the sump, which could mean that they are broken down by biogeochemical reactions that have CO$_2$, CH$_4$ and N$_2$O as by-products. In the septic, under anaerobic conditions the high concentration of organic matter in wastewater is degrading via hydrolysis, acidogenesis and methanogenesis producing both CO$_2$, CH$_4$ and NH$_4^+$. 
Table 8.2: Quality of effluent going into the ET system and in the sumps.

<table>
<thead>
<tr>
<th></th>
<th>ST effluent</th>
<th>ET 1</th>
<th>ET 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>COD (mg l^{-1})</td>
<td>583.6 ± 48.4</td>
<td>40.9 ± 9.2</td>
<td>34.6 ± 8.8</td>
</tr>
<tr>
<td>TOC (mg l^{-1})</td>
<td>258.0 ± 38.5</td>
<td>26.2 ± 8.3</td>
<td>19.9 ± 7.0</td>
</tr>
<tr>
<td>NH_4^+N (mg l^{-1})</td>
<td>74.0 ± 8.8</td>
<td>19.0 ± 14.3</td>
<td>8.9 ± 2.7</td>
</tr>
<tr>
<td>NO_2^-N (mg l^{-1})</td>
<td>0.1 ± 0.0</td>
<td>0.1 ± 0.0</td>
<td>0.1 ± 0.0</td>
</tr>
<tr>
<td>NO_3^-N (mg l^{-1})</td>
<td>0.4 ± 0.2</td>
<td>0.2 ± 0.0</td>
<td>0.2 ± 0.0</td>
</tr>
<tr>
<td>TN (mg l^{-1})</td>
<td>210.4 ± 21.4</td>
<td>46.2 ± 32.5</td>
<td>21.0 ± 5.9</td>
</tr>
<tr>
<td>PO_4^{3-}P (mg l^{-1})</td>
<td>12.8 ± 2.4</td>
<td>2.7 ± 0.6</td>
<td>1.0 ± 0.5</td>
</tr>
<tr>
<td>SO_4^{2-} (mg l^{-1})</td>
<td>53.2 ± 9.1</td>
<td>6.2 ± 1.3</td>
<td>6.8 ± 1.8</td>
</tr>
<tr>
<td>Cl (mg l^{-1})</td>
<td>102.6 ± 17.7</td>
<td>36.9 ± 17.6</td>
<td>26.9 ± 5.9</td>
</tr>
<tr>
<td>TC (l^{-1})</td>
<td>1 208 600 ± 746 445</td>
<td>7149 ± 12 224</td>
<td>13 144 ± 16 467</td>
</tr>
<tr>
<td>E. coli (l^{-1})</td>
<td>512 200 ± 283 820</td>
<td>402 ± 362</td>
<td>945 ± 991</td>
</tr>
</tbody>
</table>

### 8.4.3 CO2 fluxes

**CO2 fluxes during survey measurements** from the willow ranged from 0.21 to 0.73 µmol CO2 m^{-2} s^{-1}, from 0.19 to 0.70 µmol CO2 m^{-2} s^{-1}, from 1.09 to 5.18 µmol CO2 m^{-2} s^{-1} and from 1.14 to 48.26 µmol CO2 m^{-2} s^{-1} at Q1, Q2, Q3 and Q4, respectively. Over the control area, the CO2 flux values varied between 0.82 and 1.17 µmol CO2 m^{-2} s^{-1}, 1.27 and 1.69 µmol CO2 m^{-2} s^{-1}, 4.56 and 5.78 µmol CO2 m^{-2} s^{-1} and 6.62 and 12.31 µmol CO2 m^{-2} s^{-1}, see Figure 8.5. During Q1 (end of growing season) and Q2 (dormant season), the CO2 fluxes were in the same low range, but showing less variability during Q1. The CO2 fluxes from the control area increased in Q2 however, they exceeded the CO2 fluxes from the willow bed at both quarters. During Q3 (early growing season) and Q4 (peak growing season), CO2 fluxes show higher spatial variability over the willow bed; however, the minimum CO2 fluxes were higher at the control than at the willow bed.
Figure 8.5: Spatial interpolation of CO$_2$ fluxes over the willow bed at a – Q1 (growing season – end of October 2017), b – Q2 (end of dormant season – end of March 2018), c – Q3 (early growing season – beginning of June) and d – Q4 (peak growing season – beginning of August). Colours (from blue to red) indicate the fluxes at each quarters and the grey scale show fluxes on the overall scale.
**CO₂ fluxes during long-term measurements** from the willow bed ranged from 0.12 to 2.33 μmol CO₂ m⁻² s⁻¹, from 0.13 to 1.37 μmol CO₂ m⁻² s⁻¹, from 1.60 to 9.48 μmol CO₂ m⁻² s⁻¹ and from 1.96 to 56.65 μmol CO₂ m⁻² s⁻¹ at Q1, Q2, Q3 and Q4, respectively. Over the control area, the CO₂ flux values varied between 0.30 and 2.99 μmol CO₂ m⁻² s⁻¹, 1.11 and 2.55 μmol CO₂ m⁻² s⁻¹, 4.09 and 9.05 μmol CO₂ m⁻² s⁻¹ and 6.46 and 15.31 μmol CO₂ m⁻² s⁻¹, see Figures 8.6a - 8.6d.

The CO₂ fluxes from the willow bed were statistically significantly lower than the fluxes from the control sites which had higher mean values. The only exception was during Q1, when the highest CO₂ fluxes were measured at 15 m from the inflow and the fluxes at 7 and 25 m were not statistically significantly different from the control area: note that there was no measurements at 2 m for this quarter.

---

![Figure 8.6](image)
8.4.4 CH$_4$ fluxes

**CH$_4$ fluxes combined from survey measurements and gas sampling** from the willow bed ranged from 8.41 to 68.72 nmol CH$_4$ m$^{-2}$ s$^{-1}$, from $-1.10$ to $19.61$ nmol CH$_4$ m$^{-2}$ s$^{-1}$, from $-5.70$ to 4.93 nmol CH$_4$ m$^{-2}$ s$^{-1}$ and from $-3.74$ to 51.35 nmol CH$_4$ m$^{-2}$ s$^{-1}$ at Q1, Q2, Q3 and Q4, respectively. Flux values at control collars were in the detection limit only in Q4 ranging between $-1.71$ to $-0.36$ nmol CH$_4$ m$^{-2}$ s$^{-1}$. The highest CH$_4$ values were observed during Q1 and Q4 at 15 and 25 m, see Figure S.7. The highest CH$_4$ fluxes occurred during Q1, at the end of the growing season.

**CH$_4$ fluxes during long-term measurements** from the ET willow system ranged from $-0.76$ to 12.57 nmol CH$_4$ m$^{-2}$ s$^{-1}$ and from $-1.89$ to 4310.44 nmol CH$_4$ m$^{-2}$ s$^{-1}$ at Q1 and Q4, respectively. Over the control area, the CH$_4$ flux values varied between $-0.86$ and 0.61 nmol CH$_4$ m$^{-2}$ s$^{-1}$ and $-0.86$ and 0.44 nmol CH$_4$ m$^{-2}$ s$^{-1}$, as shown in Figures S.8a - S.8d. During Q1, CH$_4$ fluxes at 7, 15 and 25 m were statistically significantly higher than those fluxes at the control site. During Q4, the highest CH$_4$ fluxes occurred at 15 m similar to where the CO$_2$ fluxes occurred, showing really strong GHG formation there. Also much higher CH$_4$ fluxes in general during Q4 this is associated with the water emptying out of the bed after long flooded period leading to the CH$_4$ emissions. Similar uniquely high CH$_4$ flux was found from a soakaway after a long period of flooded period (Somlai et al., 2019).
Figure 8.7: Spatial interpolation of CH$_4$ fluxes over the willow bed at four different times:

- Q1 (growing season - end of October 2017)
- Q2 (end of dormant season - end of March 2018)
- Q3 (early growing season - beginning of June)
- Q4 (peak growing season - beginning of August)

The colors (from blue to red) indicate the fluxes at each quarter, with the grey scale showing overall scale.

- Q1: Fluxes are mostly low with a few exceptions.
- Q2: There is a slight increase in fluxes compared to Q1.
- Q3: Fluxes are higher than in Q2, with some significant peaks.
- Q4: Highest fluxes observed, with a clear peak in the center.

8. GHG EMISSIONS FROM WILLOW SYSTEM
8.4. RESULTS AND DISCUSSION

8.4.5 N₂O fluxes

N₂O fluxes during gas sampling from the willow bed ranged between -93.07 and 74.30 nmol N₂O m⁻² s⁻¹, -246.72 and 61.70 nmol N₂O m⁻² s⁻¹, -337.70 and 310 nmol N₂O m⁻² s⁻¹, 148.51 and 74.30 nmol N₂O m⁻² s⁻¹, at Q1, Q2, Q3 and Q4, respectively. At the control area, fluxes were outside the detection limit during the Q1, during Q2 one valid of 217.93 nmol N₂O m⁻² s⁻¹, during Q3 ranged between -135.17 and 283.99 nmol N₂O m⁻² s⁻¹ and during Q4 one valid measurement of 67.02 nmol N₂O m⁻² s⁻¹, see Figure 8.9. The highest N₂O fluxes occurred at 15 m.

During Q1 (end of growing season) and Q2 (dormant season), the N₂O fluxes were in the lower range, they were in the higher range during Q3 (early growing season) and Q4 (peak growing season).

8.4.6 Influence of environmental parameters on GHG fluxes

Investigating the discrete measurements, positive correlations were found between CO₂ surface fluxes and water temperature in the inspection well, air temperature and evapotranspiration. Negative correlations were found between CO₂ surface fluxes and water level in the inspection well and relative humidity, see Figure E.1. Using the diurnal long-term hourly measurements, daily averages of CO₂ fluxes and environmental parameters were calculated in order to find correlations, see Figure E.4. During these measurements, soil temperature and VWC was also measured at each collar. Additionally to the similar positive correlations between CO₂ surface fluxes and water temperature in the inspection...
8. GHG EMISSIONS FROM WILLOW SYSTEM

Figure 8.9: Spatial interpolation of N\textsubscript{2}O fluxes over the willow bed at a – Q1 (growing season - end of October 2017), b – Q2 (dormant season - end of March 2018), c – Q3 (early growing season - beginning of June) and d – Q4 (peak growing season - beginning of August). Colours (from blue to red) indicate the fluxes at each quarters and the grey scale show fluxes on the overall scale.
well, air temperature and evapotranspiration, positive correlations were found between the CO\textsubscript{2} fluxes and soil temperature.

There was not a sufficient amount of data to draw any conclusions on how environmental parameters are influencing CH\textsubscript{4} and N\textsubscript{2}O fluxes.

### 8.4.7 Total GHG emissions from the willow bed

Inverse Distance Weighted (IDW) interpolation of the flux values was used to estimate the total emissions from the willow bed for each different period of willow growing season (i.e. quarter), see Figure 8.5, 8.7 and 8.9. Each tile represents 0.5 m x 0.5 m area of the willow bed. These values from the tiles were summed up for each quarter and in order to provide emission estimates and these emissions from each quarters were added together to provide estimate of annual fluxes. The emission values were multiplied by two to represent the two willow ET beds and they were then normalised per person by dividing by the number of people using the wastewater system. Comparing these interpolated values with other studies on similar systems, see Table 8.3, the CH\textsubscript{4} and N\textsubscript{2}O emissions are in the range of the previous studies (although the CH\textsubscript{4} is somewhat higher in general). However, the predicted CO\textsubscript{2} emissions significantly exceeded earlier observed CO\textsubscript{2} emissions by more than a factor of 10 in most cases. This is due to the fact that total emissions calculation is based on the area of the treatment unit. Further, there is effect the presence of plants has on C and N dynamics in soil, and on the microbial processes that transform C and N. For example, plants add C to the soil via roots, roots exudates, and leaves, all of which can increase CO\textsubscript{2} flux. Processing of this material consumes O\textsubscript{2}, which may result in hypoxia or anoxia in some areas of the soil, affecting N\textsubscript{2}O and CH\textsubscript{4} dynamics. Roots also respire, producing CO\textsubscript{2}. Also some plants may emit N\textsubscript{2}O and CH\textsubscript{4} naturally.
Table 8.3: Summary of total estimated GHG emissions and flux rates from DWWTSs.

<table>
<thead>
<tr>
<th></th>
<th>CO₂ kg yr⁻¹ cap⁻¹</th>
<th>CH₄ kg yr⁻¹ cap⁻¹</th>
<th>N₂O kg yr⁻¹ cap⁻¹</th>
<th>Total emissions CO₂Eq kg yr⁻¹ cap⁻¹</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Diaz-Valbuena et al. (2011)</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>septic tank²</td>
<td>12.15 ± 0.99</td>
<td>4.02 ± 0.80</td>
<td>0.002 ± 1.570</td>
<td>113.2 ± 468.2</td>
</tr>
<tr>
<td>vent pipe²</td>
<td>122.28 ± 20.77</td>
<td>3.91 ± 0.62</td>
<td>0.073 ± 1.310</td>
<td>241.8 ± 391.2</td>
</tr>
<tr>
<td><strong>Truhlar et al. (2016)</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>vent pipe³</td>
<td>58.40 ± 1.17</td>
<td>4.02 ± 4.38</td>
<td>0.04 ± 1.40</td>
<td>170.8 ± 431.3</td>
</tr>
<tr>
<td>sand filter³</td>
<td>43.80 ± 30.30</td>
<td>0.003 ± 0.017</td>
<td>0.002 ± 0.510</td>
<td>44.5 ± 155.0</td>
</tr>
<tr>
<td>STU³</td>
<td>47.45 ± 43.80</td>
<td>-0.001 ± 0.020</td>
<td>0.008 ± 0.657</td>
<td>49.8 ± 200.6</td>
</tr>
<tr>
<td><strong>Somlai-Haase et al. (2017)</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>soakaway³</td>
<td>56.58 ± 75.92</td>
<td>-</td>
<td>-</td>
<td>56.58 ± 75.92</td>
</tr>
<tr>
<td><strong>Somlai et al. (2019) in prep</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>soakaway³</td>
<td>29.96 ± 10.57</td>
<td>-0.0001 ± 0.0420</td>
<td>-</td>
<td>29.96 ± 10.62</td>
</tr>
<tr>
<td><strong>This study</strong></td>
<td></td>
<td></td>
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<td></td>
</tr>
<tr>
<td>gross willow⁴</td>
<td>928</td>
<td>0.38</td>
<td>0.03</td>
<td>945</td>
</tr>
<tr>
<td>net willow⁵</td>
<td>246</td>
<td>0.39</td>
<td>0.01</td>
<td>258</td>
</tr>
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</table>

<table>
<thead>
<tr>
<th></th>
<th>CO₂ kg yr⁻¹ cap⁻¹ m⁻²</th>
<th>CH₄ g yr⁻¹ cap⁻¹ m⁻²</th>
<th>N₂O g yr⁻¹ cap⁻¹ m⁻²</th>
<th>Total flux rates CO₂Eq kg yr⁻¹ cap⁻¹ m⁻²</th>
</tr>
</thead>
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<tr>
<td><strong>Somlai-Haase et al. (2017)</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>soakaway³</td>
<td>9.43 ± 12.65</td>
<td>-</td>
<td>-</td>
<td>9.43 ± 12.65</td>
</tr>
<tr>
<td>Somlai-Haase in prep</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>soakaway³</td>
<td>5.00 ± 1.76</td>
<td>-0.02 ± 7.00</td>
<td>-</td>
<td>4.99 ± 1.77</td>
</tr>
<tr>
<td><strong>This study</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>gross willow⁴</td>
<td>3.76</td>
<td>2.0</td>
<td>0.1</td>
<td>3.83</td>
</tr>
<tr>
<td>net willow⁵</td>
<td>1.00</td>
<td>2.0</td>
<td>0.04</td>
<td>1.05</td>
</tr>
</tbody>
</table>

² geometric mean ± SD; ³ arithmetic mean ± SD; ⁴ median ± SD; ⁵ sum of the seasonal interpolation; ⁶ gross minus control flux.
8.5 Conclusions

GHG fluxes had strong spatial and seasonal variation from the willow bed. CO$_2$ and N$_2$O fluxes were in the lower range at the end of the growing season as well as across the dormant season, but then higher during the early growing season and the peak growing season. The highest CH$_4$ fluxes occurred at the end of the growing season. The high spatial variation could be caused by different environmental parameter for example the heterogeneity of the soil or vegetation or microtopography or shading from surrounding vegetation. The biomat formation can also influence the spatial heterogeneity of the emissions. The presence of plants has effect on C and N dynamics in soil and on the microbial processes that transform C and N. For example, plants add C to the soil via roots, root exudates, and leaves, all of which can increase CO$_2$ flux. Processing of this material consumes O$_2$, which may result in hypoxia or anoxia in some areas of the soil, affecting N$_2$O and CH$_4$ dynamics. Roots also respire, producing CO$_2$. And some plants may emit N$_2$O and CH$_4$ naturally.

The CO$_2$ fluxes had positive correlations with soil and air temperatures, evapotranspiration and negative correlation with water level and relative humidity. There was not a sufficient amount of data to draw any conclusions on how environmental parameters influenced CH$_4$ and N$_2$O fluxes. The total net GHG emissions from the evapotranspiration system (258 CO$_2$Eq. kg yr$^{-1}$ cap$^{-1}$) which exceeds measured emissions from other on-site septic systems, however; this high emission rates are accounted for the large extend of the willow bed.
8. GHG EMISSIONS FROM WILLOW SYSTEM
Conclusions and recommendations

The main aim of this research was to develop a better understanding of the contribution of the DWWTSs to the GHG inventories by quantifying and qualifying the on-site production of GHGs including CO\(_2\), CH\(_4\) and N\(_2\)O using established field research sites set up by the Department of Civil, Structural and Environmental Engineering at TCD. The GHG emission rates were quantified from two STs, a soakaway, two engineered STUs receiving different levels of wastewater pre-treatments; primary treated ST effluent versus secondary treated effluent as well as from a willow-based evapotranspiration system. The outcomes of this research are relevant for any system that discharges effluent to ground, which could include decentralised clustered systems.

9.1 Summary of findings

Over the soakaway, the CO\(_2\) emissions ranged from 25.1 to 162.9 g CO\(_2\) cap\(^{-1}\) d\(^{-1}\) (median of 82.2 g CO\(_2\) cap\(^{-1}\) d\(^{-1}\)). The CH\(_4\) emissions ranged from -0.006 to 1.32 g CH\(_4\) cap\(^{-1}\) d\(^{-1}\) (median of -0.0006 g CH\(_4\) cap\(^{-1}\) d\(^{-1}\)). No strong spatial variation of CO\(_2\) fluxes was revealed and it seems that the CO\(_2\) fluxes are strongly driven by the environmental parameters of soil temperature, volumetric water content and pore water EC. However, CH\(_4\) fluxes did show a strong spatial distribution on this site with the highest emission rate the closest to the effluent point of the ST, where 30% of the total emission was released over a 3-day period. The soakaway consumed overall 0.033 kg CO\(_2\)Eq yr\(^{-1}\) less CH\(_4\) and emitted 7.327 kg CO\(_2\) yr\(^{-1}\) more CO\(_2\) than a similarly sized area of control soil.

GHG emissions were variable from the different stages (i.e. ST and STU) of the treatment process of the DWWTSs and between the two different sites. The inclusion of up-front
rotating bio contactor had a strong impact on the net GHG emissions, however, the total net emissions were not affected strongly by the inclusion of the coconut husk filter packaged secondary treatment unit. The total net emissions from the full systems were 17.0 and 21.9 CO$_2$Eq. kg cap$^{-1}$ yr$^{-1}$ at the two different sites, respectively. Over 80% of the total net emissions was in the form of CO$_2$, around 15% in CH$_4$ and less than 2% in N$_2$O. At one of the site where rotating biofilter contactor were used as secondary treatment, significant difference was observed between net emissions comparing septic tank effluent and secondary effluent. 39.6 CO$_2$Eq. kg cap$^{-1}$ yr$^{-1}$ total net emissions were estimated from the septic tank effluent and $-5.6$ CO$_2$Eq. kg cap$^{-1}$ yr$^{-1}$ uptake from the system receiving secondary effluent. At the other site, no significant difference were observed between trenches receiving septic tank or secondary effluent of coconut husk filter.

GHG fluxes had strong spatial and seasonal variation from the willow evapotranspiration system. CO$_2$ and N$_2$O fluxes were in the lower range at the end of the growing season and at the dormant season and higher at the early growing season and peak growing season. The highest CH$_4$ fluxes occurred at the end of the growing season. The CO$_2$ fluxes had positive correlations with temperatures and evapotranspiration and negative correlations with water level and relative humidity. There was not a sufficient amount of data to draw any conclusions on how environmental parameters are influencing CH$_4$ and N$_2$O fluxes. The total gross GHG emission from the evapotranspiration system (258 kg CO$_2$Eq. yr$^{-1}$ cap$^{-1}$) exceeded measured emissions from other on-site septic systems.

Unique high CO$_2$ fluxes – up to 20–times higher than the overall median, were measured in October 2015 at the KB site from the soakaway. However, the environmental parameters recorded during the study were not able to explain these high values. One hypothesis for these uniquely high emissions is that maybe some chemical was discharged into the septic tank that killed the microorganisms forming the biomat in the soakaway.

Unique high CH$_4$ fluxes – were measured at the KB site from the soakaway in April 2016 and 30% of the total CH$_4$ emissions was released over a 3-day period. Uniquely high emissions can be caused by the 'birch effect' (Birch, 1958). 'Birch pulse' is caused by rapidly increased respiration and mineralisation rates in response to changing moisture conditions, however; only CH$_4$ emissions were extremely high and CO$_2$ reminded on normal. Nevertheless, as drying and rewetting events are forecasted to become more frequent in future with climate change (IPCC, 2013), this can lead to increased emissions rate from STUs as well.

Controlling environmental factors – while temperature, soil water content, and atmospheric pressure were identified as the most significant environmental factors correlated to the release of CO$_2$ from the control sites, fluxes from the soakaway and STUs showed
weaker correlations in regard to environmental factors. Among the recorded environmental parameters, atmospheric and soil temperature were the best predictors of CO₂ fluxes. CH₄ fluxes can not be explained with environmental drivers based on this collected data from soakaway. However, CH₄ flux rates do seem to be related to pooled anaerobic/anoxic effluent drying out.

9.2 Implication of emissions

Wastewater can be a source of CH₄ when treated or disposed anaerobically. It can also be a source of N₂O emissions. CO₂ emissions from wastewater are not considered in the 2006 IPCC guidelines because these are of biogenic origin and should not be included in national total emissions. Domestic wastewater is defined as wastewater from household water use. Domestic wastewater is either treated in centralized treatment plants or in septic tanks. Centralised wastewater treatment plants also treat commercial and industrial wastewater and for that reason emissions from Industrial Wastewater (5.D.2) are included in Domestic Wastewater (5.D.1).

Approximately two-thirds of the population in the Republic of Ireland is served by centralised sewerage treatment plants, the remaining one-third of the population uses septic tanks to treat wastewater mainly for individual houses in non-urban areas (Smith et al., 2004). There are an estimated 489,069 (CSO 2016) domestic waste water treatment systems treating waste water from single houses in Ireland that are not connected to a public sewer system which utilise conventional septic tanks. These households systems are in rural areas and therefore not connected to urban centralised wastewater treatment systems which cater for urban domestic, commercial and industrial wastewaters. National statistics on household occupancy suggest a value of approximately 3 persons per household (CSO, 2016), therefore these domestic septic tanks service approximately 1.5 million people or one third of Irelands population.

In 2016, the Irish national total GHG emissions from the wastewater treatment and discharge sector was estimated to be 147.12 kt CO₂Eq. or 30.90 kg CO₂Eq. cap⁻¹ of which 34 % is in the form of CH₄ and 66 % is in the form of N₂O (CSO, 2017; EPA, 2018a). As it was mentioned earlier, CO₂ emissions from wastewater are not considered in the IPCC (2006) guidelines because these are of biogenic origin and according to them CO₂ emissions should not be included in national total emission estimates. The Irish calculation is based on that approximately two-thirds of the population in Ireland is served by centralised sewerage treatment plants, the remaining one-third of the population uses septic tanks
to treat wastewater mainly for individual houses in non-urban areas \citep{CSO2017}. The national inventories of GHG emissions from septic systems are based on the assumption of the conversion rate of organic matter entering the systems from a limited number of direct field measurements and only assuming emissions are generated in the STs and considering STUs as negligible sources \citep{IPCC2006,USEPA2016}. In 2016, emissions from wastewater treatment and discharge account for 15.4\% of total emissions from the waste sector in Ireland. However, the contribution of this sub-category is negligible (0.24\%) in comparison to the total national GHG emissions of 61,545.82 kt CO$_2$Eq. or 12.9 t CO$_2$Eq. cap$^{-1}$ yr$^{-1}$ \citep{EPA2018a}.

During this PhD project, the measured net GHG emissions from DWWTSs ranged between 3.7 and 258 kg CO$_2$Eq. yr$^{-1}$ cap$^{-1}$ including CO$_2$ emissions, furthermore, for all systems the CO$_2$ emissions were over 80\% of the total emissions. However, the national estimates exclude direct CO$_2$ emissions from wastewater. Account only for CH$_4$ and N$_2$O emissions from DWWTSs in this study, the emissions ranged between −0.01 and 3.5 kg CO$_2$Eq cap$^{-1}$ yr$^{-1}$. These values are much lower than the national estimate of GHG emissions from the wastewater treatment and discharge sector, 30.90 kg CO$_2$Eq cap$^{-1}$ yr$^{-1}$. Further direct field measurements are needed to revised the national estimate of GHG emissions from the wastewater treatment and discharge sector for domestic wastewater.

### 9.2.1 Recommendations for further research

During the course of this study a number of areas for further research were identified, as summarised below:

- Microbial communities are the fundamental driver for the biochemical cycles which emit GHGs. Hence, sampling campaign of microbial communities in the soil across the CC and KM STUs was carried out on the 16$^{th}$ and 17$^{th}$ August 2018 at CC and KM site with collaboration of University of Newcastle with Carolina Ospina Betancourth and Tom Curtis. The collected samples will be analysed for metagenomics (culture-independent genomic analysis) and screened for 16S rRNA genes. Unfortunately, due to the timing of the field campaign these results have not been processed yet and so cannot be included into this thesis. However, linking the GHG emissions to the different microbiological communities is an area which warrants further detailed studies.

- The soil biomass growth in the percolation trenches were studied at KM and CC by another PhD study running concurrently to this research. This will produce a
detailed investigation on the relationship between the biomass and GHG. We assume that where the biomat developed we will find higher emissions of GHGs as there are more available C.

- As collecting samples and analysing for N$_2$O emissions was really time-consuming and the departmental instrument was not sensitive to detect the lower concentration, it would be interesting to do on-site measurements with a more precise N$_2$O gas analyser. I would recommend to use on-site analyser for N$_2$O fluxes, for example G2308 or G2508 Gas Concentration Analyser by Picarro.
Appendix A

Technical specifications of greenhouse gas measurements
### APPENDIX A. AUTOMATED CHAMBER SYSTEM

<table>
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<tr>
<th>Chamber</th>
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<tr>
<td>Dimensions</td>
<td>48.3 cm L x 38.1 cm W x 33.0 cm H</td>
</tr>
<tr>
<td>Weight</td>
<td>5.9 kg</td>
</tr>
<tr>
<td>Headspace volume</td>
<td>4076.1 cm³</td>
</tr>
<tr>
<td>Dome</td>
<td>opaque round</td>
</tr>
<tr>
<td>Material</td>
<td>white coated stainless steel</td>
</tr>
<tr>
<td>Sealing</td>
<td>1 neoprene gasket plus 1 neoprene collar gasket, black</td>
</tr>
<tr>
<td>Vent</td>
<td>special vent design and no fan</td>
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<td>Dimensions</td>
<td>20.3 cm I.D./ 21.3 cm O.D.</td>
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<tr>
<td>Enclosed soil area</td>
<td>318 cm²</td>
</tr>
<tr>
<td>Insertion depth and offset</td>
<td>≈10 cm and ≈5 cm</td>
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<td>Material</td>
<td>PVC</td>
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<tbody>
<tr>
<td>Dimensions</td>
<td>15 m L x 3.2 mm I.D.</td>
</tr>
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<td>Material</td>
<td>Bev-a-line, protected inside a black plastic tube</td>
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<td>Weight</td>
<td>9.4 kg</td>
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<tr>
<td>Pump</td>
<td>Diaphragm</td>
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<tr>
<td>Flow rate</td>
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<td>Principle</td>
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<tr>
<td>Measurement range</td>
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<tr>
<td></td>
<td>0-40 mol mol⁻¹ H₂O</td>
</tr>
<tr>
<td>Uncertainty</td>
<td>accuracy: 1.5% of reading</td>
</tr>
<tr>
<td></td>
<td>RMS noise &lt;1 ppm CO₂⁴</td>
</tr>
<tr>
<td></td>
<td>RMS noise &lt;0.01 mol mol⁻¹ H₂O⁵</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Los Gatos</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Principle</td>
<td>Laser Absorption Spectroscopy</td>
</tr>
<tr>
<td>Measurement range</td>
<td>0.01-100 ppm CH₄</td>
</tr>
<tr>
<td></td>
<td>1- 20000 ppm CO₂</td>
</tr>
<tr>
<td>Total gas volume</td>
<td>5372- 6294 m³</td>
</tr>
<tr>
<td>Temperature range</td>
<td>-20 to ±45 °C</td>
</tr>
<tr>
<td>Operating Humidity range</td>
<td>0 to 95% RH, non-condensing</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Accessories</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Air temperature</td>
<td>thermistor, accuracy ±0.5°C</td>
</tr>
<tr>
<td>Soil temperature</td>
<td>thermistor, accuracy ±1.0°C</td>
</tr>
<tr>
<td>Soil moisture</td>
<td>Decagon ECH₂O model EC-5, ±3 % VWC</td>
</tr>
<tr>
<td>Air pressure</td>
<td>1.5 % accuracy</td>
</tr>
<tr>
<td>Power requirement</td>
<td>max 60 W</td>
</tr>
</tbody>
</table>

---

⁰ (Parkin and Venicrea 2010); ⁱ chamber to/from multiplexer; ⁲ multiplexer to gas analyser; ⁴ at 370 ppm with 1 s signal averaging; ⁵ at 10 ppt with 1 s signal averaging
<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>GC</strong></td>
<td></td>
</tr>
<tr>
<td>Sampling rate</td>
<td>6.25 pts per s</td>
</tr>
<tr>
<td>Column type</td>
<td>Elite PLOT Q</td>
</tr>
<tr>
<td>Column dimensions</td>
<td>2 x 30 m x 0.53 mm</td>
</tr>
<tr>
<td>Carrier gas</td>
<td>N$_2$ 5 psi</td>
</tr>
<tr>
<td>Valve temperature</td>
<td>90°C</td>
</tr>
<tr>
<td>Oven temperature</td>
<td>35°C</td>
</tr>
<tr>
<td>Run time</td>
<td>4 min</td>
</tr>
<tr>
<td>Split</td>
<td>5 ml min$^{-1}$</td>
</tr>
<tr>
<td><strong>FID</strong></td>
<td></td>
</tr>
<tr>
<td>Temperature</td>
<td>200°C</td>
</tr>
<tr>
<td>H$_2$ flow</td>
<td>45.0 ml min$^{-1}$</td>
</tr>
<tr>
<td>Air flow</td>
<td>450 ml min$^{-1}$</td>
</tr>
<tr>
<td>CH$_4$ peak time</td>
<td>3.32 min</td>
</tr>
<tr>
<td><strong>ECD</strong></td>
<td></td>
</tr>
<tr>
<td>Temperature</td>
<td>350°C</td>
</tr>
<tr>
<td>Make-up gas flow</td>
<td>30.0 ml min$^{-1}$</td>
</tr>
<tr>
<td>N$_2$O peak time</td>
<td>3.49 min</td>
</tr>
</tbody>
</table>
Appendix B

Technical specifications of auxiliary probes
Table B.1: Technical specifications of digital pH, dissolved $O_2$, EC and temperature probe WTW GmbH, Germany.

<table>
<thead>
<tr>
<th>Probe</th>
<th>Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH SenTix 980</td>
<td>Reference electrolyte: KCl 3 mol/L, Ag+ free</td>
</tr>
<tr>
<td></td>
<td>Junction: Ceramic</td>
</tr>
<tr>
<td></td>
<td>pH measuring range: 0.000 – 14.000</td>
</tr>
<tr>
<td></td>
<td>Temperature range: 0 to 80°C</td>
</tr>
<tr>
<td>EC TetraCon 925</td>
<td>Measuring principle: Four-electrodes measurement</td>
</tr>
<tr>
<td></td>
<td>Cell constant: 0.475 cm$^{-1}$ ±1.5 %</td>
</tr>
<tr>
<td></td>
<td>Temperature sensor: Integrated NTC (30Kohm at 25°C)</td>
</tr>
<tr>
<td></td>
<td>Temperature range: 0 to 80°C</td>
</tr>
<tr>
<td></td>
<td>Dimensions: 176 mm x 21.7 mm</td>
</tr>
<tr>
<td></td>
<td>Weight: ≈60 g (without cable)</td>
</tr>
<tr>
<td>FDO 925</td>
<td>Measuring principle: Optical measurement based on photoluminescence</td>
</tr>
<tr>
<td></td>
<td>Weight: 180 g</td>
</tr>
<tr>
<td></td>
<td>Temperature sensor: Integrated NTC (30Kohm at 25°C)</td>
</tr>
<tr>
<td></td>
<td>Dimensions: 205.5 mm x 21.7 mm</td>
</tr>
<tr>
<td></td>
<td>$O_2$ measuring range: 0 to 20 mg L$^{-1}$ D.O.</td>
</tr>
<tr>
<td></td>
<td>Temperature range: 0 to 80°C</td>
</tr>
<tr>
<td></td>
<td>Accuracy: ±1.5 %</td>
</tr>
</tbody>
</table>
Table B.2: Technical specifications of hot wire anemometer, LU8050, TQC Sheen.

<table>
<thead>
<tr>
<th>Specification</th>
<th>Details</th>
</tr>
</thead>
<tbody>
<tr>
<td>Operating temperature</td>
<td>0 to 50°C</td>
</tr>
<tr>
<td>Humidity</td>
<td>less than 80% RH</td>
</tr>
<tr>
<td>Power supply</td>
<td>9V battery</td>
</tr>
<tr>
<td>Power current</td>
<td>DC 60 90mA</td>
</tr>
<tr>
<td>Weight</td>
<td>280 g</td>
</tr>
<tr>
<td>Dimensions</td>
<td>210 mm x 75 mm x 50 mm</td>
</tr>
<tr>
<td>Air velocity</td>
<td>0.1 - 25.0 m/s</td>
</tr>
<tr>
<td>Resolution</td>
<td>0.01 m/s</td>
</tr>
<tr>
<td>Accuracy</td>
<td>±5 %</td>
</tr>
<tr>
<td>Temperature Range</td>
<td>0 to 50°C</td>
</tr>
<tr>
<td>Resolution</td>
<td>0.1 °C</td>
</tr>
<tr>
<td>Accuracy</td>
<td>±1 °C</td>
</tr>
</tbody>
</table>
Table B.3: Data recorded with automated weather station.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Unit</th>
<th>Instrument</th>
<th>Aggregation</th>
<th>Log interval</th>
</tr>
</thead>
<tbody>
<tr>
<td>case temperature</td>
<td>$T_{\text{case}}$</td>
<td>°C</td>
<td>Type T Thermocouple</td>
<td>sample</td>
<td>H</td>
</tr>
<tr>
<td>air temperature</td>
<td>$T_{\text{air}}$</td>
<td>°C</td>
<td>Campbell CS215</td>
<td>mean, min, max</td>
<td>H-T</td>
</tr>
<tr>
<td>relative humidity</td>
<td>$U_{\text{rel}}$</td>
<td>–</td>
<td>Campbell CS215</td>
<td>mean, min, max</td>
<td>H</td>
</tr>
<tr>
<td>atmospheric pressure</td>
<td>$\rho$</td>
<td>mbar</td>
<td>Vaisala PTB110</td>
<td>sample</td>
<td>H</td>
</tr>
<tr>
<td>rain fall</td>
<td>$r$</td>
<td>mm</td>
<td>EML ARG100</td>
<td>total</td>
<td>H-T-M</td>
</tr>
<tr>
<td>wind speed</td>
<td>$u_2$</td>
<td>m s$^{-1}$</td>
<td>Young 05103-5</td>
<td>mean</td>
<td>H-T</td>
</tr>
<tr>
<td>wind gust speed</td>
<td>$u_{\text{gust}}$</td>
<td>m s$^{-1}$</td>
<td>Young 05103-5</td>
<td>max</td>
<td>H</td>
</tr>
<tr>
<td>wind direction</td>
<td>$\omega$</td>
<td>°</td>
<td>Young 05103-5</td>
<td>mean</td>
<td>H-T</td>
</tr>
<tr>
<td>net radiation</td>
<td>$R_{\text{net}}$</td>
<td>W m$^{-2}$</td>
<td>Kipp &amp; Zonen NR-Lite 2</td>
<td>mean</td>
<td>H</td>
</tr>
<tr>
<td>wastewater flow</td>
<td>$Q_{\text{SE/PE}}$</td>
<td>L</td>
<td>reed switch tipping bucket</td>
<td>total</td>
<td>H-T-M</td>
</tr>
<tr>
<td>soil volumetric water content</td>
<td>$\theta$</td>
<td>–</td>
<td>Decagon EC5/GS3</td>
<td>sample</td>
<td>H(-T-M)$^f$</td>
</tr>
<tr>
<td>soil temperature</td>
<td>$T_{\text{soil}}$</td>
<td>°C</td>
<td>Decagon GS3</td>
<td>sample</td>
<td>H-T</td>
</tr>
<tr>
<td>soil electric conductivity</td>
<td>$\kappa$</td>
<td>µS m$^{-1}$</td>
<td>Decagon GS3</td>
<td>sample</td>
<td>H(-T-M)$^f$</td>
</tr>
</tbody>
</table>

$^a$ sample – sample at time of logging; mean – mean of 1-minute interval samples (1-second intervals for wind speed, wind direction, wind gust); min – minimum of 1-minute interval samples; max – maximum of 1-minute interval samples; total – sum of additive readings per log interval.

$^b$ H – hourly interval; T – 10-minute interval; M – one-minute interval; $^c$ used as approximation for $T_{\text{air}}$ in case of T/RH probe failure; $^d$ measured as maximum average wind speed over 3-second interval; $^e$ as inflow into primary (PE) and secondary (SE) trenches; $^f$ hourly logging for EC5 sensors only
B.0.1 Suction cup lysimeters

Suction cup lysimeters were installed during site construction to extract pore water from the unsaturated zone beneath the infiltrative surface of the STU. Model 1900 Soil Water Sampler with Z1900-200 Stopper Assembly (Soilmoisture Equipment Corp., USA) were used that consists of a hollow 4.8 cm outside diameter PVC tube with a porous ceramic cup at the bottom and a TPV stopper with neoprene tubing at the top. The ceramic cup was designed with an average pore radius of 1.3 m and an air entry value of 200 kPa.

For sample collection, a suction of 50 kPa was applied using a vacuum-pressure hand pump (Model 1900K3 Extraction Kit, Soilmoisture Equipment Corp., USA). The applied suction was well below the air entry value of the ceramic cup, but created a sufficient pressure gradient between the inside of the lysimeter and the surrounding soil pores so that cohesively held pore water can migrate through the ceramic cup towards the inside of the lysimeter. The ceramic cup acted, thus, as a local sink while under suction and collected the freely moving pore water as it trickled from the infiltrative surface through the vadose zone towards the saturated zone. The tubing of the stopper assembly was, then, sealed with a clamping ring to maintain suction throughout the sampling period of one or two days.

For sample extraction from the lysimeter, the clamping ring was removed and the collected pore water was extracted using a plastic tube inserted to the bottom of the lysimeter connected to a 1 L conical flask and a vacuum-pressure hand pump (Model 1900K3 Extraction Kit, Soilmoisture Equipment Corp., USA).

A total of 53 and 52 lysimeters were successfully installed in the STU in KM (Figure B.1b) and CC (Figure B.1a), respectively; 27 and 25 in trenches receiving primary effluent, and 26 and 27 in trenches receiving secondary effluent. The lysimeters were left in the soil during the whole study to avoid disturbance of the subsoil.

Lysimeter positions were chosen in order to gain an understanding of wastewater infiltration patterns and its biogeochemical transformation processes close to the infiltrative surface (up to 55 cm depth) along the complete length of the trenches.

Before insertion, the ceramic cup was dipped into a slurry made from tap water and sieved soil (mesh size 5 mm) extracted from the hole to ensure tight contact between the ceramic cup and the surrounding soil so that pore water can move freely from the soil pores through the ceramic cup into the lysimeter. After insertion, the hole was back filled first with slurry and then the top with a layer of bentonite clay was placed around the
tube at the surface to prevent preferential flow of precipitation and ponding surface water along the tube into the unsaturated zone.

For sample collection, a suction of 50 kPa was applied using a vacuum-pressure hand pump (Model 2005G2, Soilmoisture Equipment Corp., USA;). The applied suction was well below the air entry value of the ceramic cup, but created a sufficient pressure gradient between the inside of the lysimeter and the surrounding soil pores so that cohesively held pore water can migrate through the ceramic cup towards the inside of the lysimeter. The ceramic cup acted, thus, as a local sink while under suction and collected the freely moving pore water as it trickled from the infiltrative surface through the vadose zone towards the saturated zone. The tubing of the stopper assembly was, then, sealed with a clamping ring to maintain suction throughout the sampling period of one or two days.
Figure B.1: Positions and corresponding ID numbers of lysimeters installed.
B.0.2 Soil sensors

Two types of soil sensors, type EC5 and type GS3 (METER Group Inc., USA, former Decagon Devices, Inc., USA) were installed below the infiltrative surface of the percolation trenches and outside the STU on each site to measure changes in volumetric water content, soil temperature, and electrical conductivity over time at CC and KM during site construction.

The EC5 is a two-prong capacitive sensor that determines the relative dielectric permittivity $\epsilon_r [-]$ of the soil by measuring the charge time of an hypothetical capacitor. The two plastic prongs act as electrodes and the surrounding soil as the dielectric of the capacitor. As $\epsilon_r$ varies widely across dry and saturated soil components ($\epsilon_r \approx 2 - 5$ for dry soil and $\epsilon_r \approx 81$ for water), it can be related to the volumetric soil water content $\theta$ (Topp et al., 2007) using a soil-specific calibration curve for optimal accuracy as recommended by the manufacturer (Decagon, 2015) and several studies on capacitive soil moisture measurement techniques (Evett et al., 2006; Mittelbach et al., 2011, 2012; Vaz et al., 2013).

The GS3-type soil sensor is made of a ruggedized epoxy body and three stainless steel prongs which makes this sensor more durable and easier to install in the field. Due to considerably higher costs for these sensors, only four sensors of type GS3 were place inside the STU per site: at 2 m and 12.5 m along the trenches receiving PE and SE, respectively. Apart from measuring volumetric water content of the soil, the GS3 also measures soil temperature and electrical conductivity of the pore water.
Table B.4: Technical specifications of GS3 Sensor.

<table>
<thead>
<tr>
<th>General specifications</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Dimensions</td>
<td>9.3 cm L x 2.4 cm W x 6.5 cm H</td>
</tr>
<tr>
<td>Prong length</td>
<td>5.5 cm</td>
</tr>
<tr>
<td>Cable length</td>
<td>5 m</td>
</tr>
<tr>
<td>Dielectric measurement frequency</td>
<td>70 MHz</td>
</tr>
<tr>
<td>Measurement time</td>
<td>150 ms</td>
</tr>
<tr>
<td>Power requirement</td>
<td>3.6 to 15 VDC</td>
</tr>
<tr>
<td>Operating temperature</td>
<td>−40 to 60 °C</td>
</tr>
</tbody>
</table>

| Volumetric Water Content           |                                                      |
| Accuracy                           | ±0.03 m³ m⁻³                                        |
| Resolution                         | 0.002 m³ m⁻³                                        |
| Range                              | 0.0 to 1.0 m³ m⁻³                                   |
| Volume of influence                | 160 ml                                               |

| Bulk Electrical Conductivity       |                                                      |
| Accuracy                           | ±5 %                                                 |
| Resolution                         | 0.001 dS m⁻¹                                        |
| Range                              | 0 to 25 dS m⁻¹                                       |

| Temperature                        |                                                      |
| Accuracy                           | ±1 °C                                                |
| Resolution                         | 0.1 °C                                               |
| Range                              | −40 to 60 °C                                        |
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APPENDIX B. TECHNICAL SPECIFICATIONS OF AUXILIARY PROBES

Depth below infiltrative surface [cm]

Trench 1
5
10
15

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●

●

●

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Trench 2
5
10
15

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Trench 3
5
10
15

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Trench 4
5
10
15

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●

●

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●

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●

●

1

●

●

●

●

2

3

4

5

●

●

●

●

●

●

6

7

8

9

10

11

12

13

14

15

16

17

18

Distance along trench [m]
Sensor Type

●

EC5

GS3

(a) CC

Depth below infiltrative surface [cm]

Trench 1
5
10
15

●

●

●

●

●

●

●

●

●

●

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●

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●

●

Trench 2
5
10
15

●

●

●

●

●

●

●

●

●

●

●

●

●

●

●

●

Trench 3
5
10
15

●

●

●

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●

●

●

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●

●

●

Trench 4
5
10
15

●

●

●

●

●

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●

●

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●

●

●

●

●

●

●

1

2

3

4

5

6

7

8

9

10

11

12

13

14

15

16

17

18

Distance along trench [m]
Sensor Type

●

EC5

GS3

(b) KM

Figure B.2: Positions of EC5–type (circles) and GS3–type (triangles) soil sensors installed
beneath the infiltrative surface of each trench of the STU in Crecora (a) and Kilmallock
(b).


B.1 Effluent sampling

B.1.1 Laboratory Analysis

Water samples were collected for further lab analysis, measuring organics (COD, BOD, TOC), nitrogen ($\text{NO}_3^-$, $\text{NO}_2^-$, $\text{NH}_4^+$, TON), phosphorus ($\text{PO}_4^{3-}$), coliforms and chloride. All laboratory analysis were conducted in the Analytical Laboratory at the Department of Civil, Structural and Environmental Engineering at Trinity College Dublin. Sample analysis commenced within 12 h of sampling.

COD

Merk test kit was used for measuring chemical oxygen demand (COD). Method linearity was tested for LO (10 to 1250 mg L$^{-1}$) and HI (25 to 1500 mg L$^{-1}$).

TC, TOC and TN

Total carbon (TC), total organic carbon (TOC) as non-purgeable organic carbon (NPOC) and total nitrogen (TN) were measured with a Shimadzu TOC-V CSN analyser (Shimadzu Scientific Instruments Inc., USA). For detection of TC and NPOC, a high temperature catalytic oxidation tube was used with subsequent detection of the remaining CO$_2$ in a non-dispersive infrared gas analyser (ND-IRGA). Since TOC is not directly accessible with existing analytical techniques, the organic carbon remaining in an acidified sample (using HCl to bring the sample to pH < 2) after purging the sample with carbon-free air before injection into the catalytic tube referred to as NPOC is reported instead.

For TN, combustion tube converted sample TN to nitrogen monoxide (NO) (thermal decomposition in the TC combustion tube) which was, subsequently detected in a separate chemiluminescence gas analyser (CSN module) where NO reacts with ozone (O$_3$) and converts to excited nitrogen dioxide (NO$_2$·). As the NO$_2$· returns to the ground state

<table>
<thead>
<tr>
<th>Soil component</th>
<th>Typical relative permittivity $\epsilon_r$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Air</td>
<td>1</td>
</tr>
<tr>
<td>Soil minerals</td>
<td>3 - 16</td>
</tr>
<tr>
<td>Organic matter</td>
<td>2 - 5</td>
</tr>
<tr>
<td>Water</td>
<td>80</td>
</tr>
</tbody>
</table>

Table B.5: Typical relative permittivity $\epsilon_r$ of soil components.
it emits photons, which are detected photoelectrically within the gas analyser. The area under the time integrated detector signal was related to the TN concentration in the sample using calibration curves run on serial dilutions of NaNO₃ stock solution as standard.

After reaching the laboratory, aliquots of effluent and lysimeter sample supernatant were transferred by pouring approx. 20 ml into clean 50 ml centrifuge tubes (Lennox Laboratory Supplies, the Republic of Ireland) and analysed in batches of 16 samples using a set of two OCT-L 8-port samplers (Shimadzu Scientific Instruments Inc., USA) to sequentially feed the individual samples into the analyser. ND-IRGA peak area counts were related to concentrations via calibration curves run on serial dilutions of KHP stock solution as standard.

\( \text{PO}_4^{3-}, \text{NH}_4^+, \text{TON}, \text{NO}_2^-, \text{NO}_3^-, \text{Cl}^- \text{ and SO}_4^{2-} \)

Phosphate (\( \text{PO}_4^{3-} \)), ammonium (\( \text{NH}_4^+ \)), total oxidised nitrogen (TON), nitrite (\( \text{NO}_2^- \)), chloride (\( \text{Cl}^- \)) and sulphate (\( \text{SO}_4^{2-} \)) concentrations in both effluent and lysimeter samples were determined in an automated Konelab 20i Chemistry Analyser (Thermo Fisher Scientific, USA) using the spectrophotometric phenate method (APHA, 2012, method 4500-P E) with pre-mixed reagents and standard solutions for calibration (Serosep Ltd., the Republic of Ireland).

The method detection limits were between 0.4 and 50 mg P l\(^{-1}\), 0.2 and 300 mg N l\(^{-1}\), 1.2 and 100 mg N l\(^{-1}\), 0.1 and 40 mg N l\(^{-1}\), 0.35 and 10 000 mg l\(^{-1}\) and 40 and 150 mg l\(^{-1}\) for \( \text{PO}_4^{3-}, \text{NH}_4^+, \text{TON}, \text{NO}_2^-, \text{Cl}^- \text{ and SO}_4^{2-} \), respectively.

\( \text{NO}_3^- \) concentrations were indirectly determined by subtracting measured nitrite from TON concentrations in the respective sample.

**Coliform bacteria**

Total coliform and *Escheria coli* counts were analysed using the Colilert-18 test kit (IDEXX Laboratories Inc., USA) that uses a statistical aggregation of absence/presence tests for both bacteria types simultaneously to derive most probable numbers (MPN) of colony forming units (CFU).

For the test, a substrate powder was added to 100 mL of sample collected in a sterile vessel. Serial dilutions of \( 10^{-2}, 10^{-4} \) and \( 10^{-6} \) were prepared in the laboratory for selected samples to cover expected bacterial count ranging up to \( 10^9 \) MPN 100 ml\(^{-1}\). The sample is then mixed, transferred to a tray which separates it into 97 compartments, and incubated
at 35°C for 18 h. During the incubation period the bacteria metabolised the substrate and releases either a yellow or fluorescent metabolite which marks the presence of at least one bacterium of Total coliforma or Escheria coli in the compartment, respectively. Bacterial counts expressed as MPN 100 ml$^{-1}$ were attained by enumerating the yellow and fluorescent compartments per tray.
Appendix C

Schematic drawing of collars distribution over soakaway and STU

Figure C.1: Locations of the collars at KB site. EF represent the effluent of the soakaway, along a straight line 4 collars where placed with one meter distance downhill from the effluent (KB_1, KB_2, KB_3, KB_4). KB_5 and KB_6 collars were deployed one meter right and left from the original location of KB_3 and KB_4.
Figure C.2: Locations of the collars at KM and CC. Light gray areas represent the location of the perforated pipes where the wastewater released to the trenches. The dark gray squares are the collars distributed over the trenches.
Figure C.3: Locations of collars at LO.
Appendix D

Supporting Information - Crecora and Kilmallock

D.1 Supplementary description of materials and methods

D.1.1 Flux from the ST and STU

Soil CO\textsubscript{2} and CH\textsubscript{4} fluxes were calculated using SoilFluxPro 4.0 software (Li-Cor Biosciences, Inc.).

1. CH\textsubscript{4} measurements were imported into SoilFluxPro 4.0 software (Li-Cor Biosciences, Inc.) with the appropriate time difference.

2. The start and end time of the flux analysis was set separately for CO\textsubscript{2} (30 s and 120 s) and for CH\textsubscript{4} (50 s and 350 s) in order to avoid under or overestimation of fluxes due to the different levels of fluxes and their different gas diffusion in the soil.

3. Linear and exponential lines were fit to gas concentration versus time by the software.

4. R\textsuperscript{2}-value of the regressions were used as quality control and either the linear or the exponential fit was used depending on which yielded the higher R\textsuperscript{2}-value (CO\textsubscript{2} R\textsuperscript{2} < 0.9, CH\textsubscript{4} R\textsuperscript{2} < 0.8). It should be noted that, during periods of CH\textsubscript{4} uptake, when the CH\textsubscript{4} concentration decreased over time only the linear fit was calculated. Measurements not matching this quality control were discarded from further analysis.
For discreet soil flux measurements, 99 of the CO$_2$ regressions had an R$^2$-value of 0.9 or above and 24.5% of CH$_4$ regressions had an R$^2$-value of 0.8 or above. Low CH$_4$ fluxes between $-0.3$ and 0.3 nmol CH$_4$ m$^{-2}$ s$^{-1}$ are under the detection limit of the set-up and all measurements had R$^2$-value below 0. These flux values were set at 0 for further statistics.

5. Potential CH$_4$ flux measurements were also excluded, where the initial CH$_4$ concentration was exceptional high (significantly exceeding the average atmospheric CH$_4$ concentration, above 250 ppb) and resulting unrealistic high CH$_4$ uptake.

6. The fluxes were calculated based on a mass balance approach

\[
F = \frac{V_{sys} p_0 (1 - \chi_w)}{R s T_0} \cdot \frac{\partial \chi_c(t)}{\partial t} \tag{D.1}
\]

using the total system volume $V_{sys}$ (0.06 m$^3$), the atmospheric pressure at the beginning of the measurement $p_0$ (Pa), the chamber air water vapour mole fraction $\chi_w$ (mol mol$^{-1}$), the soil collar surface area $s$ (0.032 m$^2$), the universal gas constant $R$ (8.314 Pa m$^3$ K$^{-1}$ mol$^{-1}$), the absolute temperature at the beginning of the measurement $T_0$ (K), and the initial change of chamber water vapor corrected CO$_2$ mole fraction $\partial \chi_c/\partial t$ (µmol mol$^{-1}$ s$^{-1}$) \cite{LCCOR2012}.

### D.1.2 Manual gas samples

For the manual gas samples the fluxes were calculated in excel.

1. The output of gas chromatograph; the area of the peaks representative for each gas, has linear relationship with concentration. With known standards the peak areas were converted to concentrations.

2. Data point were discarded if the GC measurements were interrupted or should malfunction.

3. For each flux measurement event, linear regression was fit to the four measured concentrations versus time.

4. R$^2$-value of the linear regression was used as quality control ($R^2 < 0.7$).

5. The fluxes were calculated based on a mass balance approach

\[
F = \frac{V_{sys} p_0 (1 - \chi_w)}{R s T_0} \cdot \frac{\partial \chi_c(t)}{\partial t} \tag{D.2}
\]
D.2. GHG EMISSIONS

using the total system volume \( V_{\text{sys}} \) (0.06 m\(^3\)), the atmospheric pressure at the beginning of the measurement \( p_0 \) (Pa), the chamber air water vapour mole fraction \( \chi_w \) (mol mol\(^{-1}\)), the soil collar surface area \( s \) (0.032 m\(^2\)), the universal gas constant \( R \) (8.314 Pa m\(^3\) K\(^{-1}\) mol\(^{-1}\)), the absolute temperature at the beginning of the measurement \( T_0 \) (K), and the initial change of chamber water vapor corrected CO\(_2\) mole fraction \( \partial \chi_c/\partial t \) (\( \mu \)mol mol\(^{-1}\) s\(^{-1}\)) (LI-COR, 2012).

6. All further data analysis and statistical analysis have being performed with R (Team, 2014).

D.1.3 Temperature correction of flux measurements

Some of the temperature sensors had a bad readings during deployments due to the failure of thermistor inside the chamber and since the accurate flux estimation depends on good chamber temperature reading, these flux values needed to be corrected following:

\[
F_{\text{corr}} = \frac{F(T_{\text{cham}} + 273.15)}{T_{\text{corr}} + 273.15} \quad \text{(D.3)}
\]

where \( F_c \) is obtained flux \( T_{\text{cham}} \) is the bad chamber temperature. \( T_{\text{corr}} \) is the right temperature from the weather station.

D.2 GHG emissions

To compare obtained flux values with previous studies, \( F_c \) was converted to per capita mass emission rates \( E_{\text{cap}} \) [g CO\(_2\) cap\(^{-1}\) d\(^{-1}\)]

\[
E_{\text{cap}} = \frac{A_{\text{soak}} M_{\text{CO}_2}}{n} \cdot F_c \quad \text{(D.4)}
\]

assuming a spatially uniform flux distribution and using the number of occupants \( n \) in the household, the surface area of the soakaway \( A_{\text{soak}} \), and the molar mass \( M_{\text{CO}_2} \) of CO\(_2\) [44.01 g mol\(^{-1}\)] as normalization factors.

Fluxes measured from the control lawn were scaled using an area equivalent to the STU (also taking an average of the control fluxes).
D.2.1 Contaminant attenuation in STU

A total of 1456 samples were collected from suction cup lysimeters in Crecora and analysed for bulk organic contaminants, nutrients, and microbial indicator organisms. Effects of dilution on measured pore water concentrations through rainfall were adjusted for by sampling campaign specific dilution correction factors taking into account the biomat growth adjusted ratio of total volume of effluent fed into the STU trenches during a 48 h period preceding the sampling event and the total volume of effective rainfall onto the STU trenches during the same time.

The results of the analysis were spatially and temporally aggregated in order to help detecting long-term trends and areas of enhanced or limited pollutant attenuation within the STU even for periods when not all installed lysimeters yielded effluent during extraction.

D.3 Results

D.3.1 ST

EC, O₂, pH and temperature in ST
Figure D.1: Water chemistry (a) electric conductivity, (b) temperature, (c) pH and (d) dissolved $\text{O}_2$ in the ST at CC and KM.

Diurnal flux measurements in the ST
Figure D.2: **A** Diurnal variation of CO$_2$ fluxes and **B** CH$_4$ from the second chamber of ST at KM and CC. The dashed lines indicate the minimum and maximum fluxes measured during the discrete sampling.

### D.3.2 STU
Figure D.3: A Spatial variation of CO$_2$ fluxes and B CH$_4$ from STU at KM and CC.
Figure D.4: Hourly mean CO$_2$ fluxes of the long-term measurements CC (a) and KM (b). Pink, blue and green colours indicate control, trenches receiving primary effluent and trenches receiving secondary effluent.
Figure D.5: Hourly mean CH$_4$ fluxes of the long-term measurements in CC (a) and KM (b). Pink, blue and green colours indicate control, trenches receiving primary effluent (PE) and trenches receiving secondary effluent (SE).
D.3.3 Effluent quantity and quality

D.3.4 Meteorological monitoring

A summary of meteorological data can be found in Figure D.8. The two sites were in relatively close proximity to each other (distance approximately 35km), the observed weather conditions were similar. Slight deviations were found in soil temperature, net radiation, and subsequently determined evaporation data as the location of the weather station in KM might have been affected by the presence of potential shading effects due to tall trees in proximity to the weather station.

As the sites were located in moderately to poorly draining soil, overall rainfall patterns affected the operation of the sites dramatically, see in Figure D.11. The summer 2018 was marked as a period of extended dry conditions with a period of more than 6 wk of nearly no effective rainfall.
D.3. RESULTS

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(a) Nonbiodegradable COD

(b) Nonbiodegradable TC

(c) Nonbiodegradable TOC

Figure D.7: Nonbiodegradable fractions for (a) COD, (b) TC, and (c) TOC for primary effluent from Crecroa (CC) and Kilmallock (KM).
Hydrological effective rainfall reaching to deeper subsoil layers was significantly limited in the summer months (April to July).

To assess effects of effluent dilution within the STU through precipitation, the effective rainfall reaching the subsoil after passing through the root zone is determined using the soil moisture deficit (SMD) model developed by Schulte et al. (2005). The SMD is defined as the amount of water (commonly expressed as mm precipitation) required to bring soil water content up to field-capacity, i.e. when the effect of gravity causes drainage in the
Figure D.10: Effective daily precipitation as sum of total rainfall and evapotranspiration over time for CC and KM.

soil profile (Keane, 2001). During periods of heavy rainfall and low evapotranspiration higher levels of effective rainfall recharge result in a greater dilution of percolating effluent within a STU compared to dry periods.

Figure D.11: Soil moisture deficit (SMD) and hydrologically effective rainfall (HER) for CC and KM. Coloured lines represent values derived from data measured on-site, grey lines represent values calculated from data obtained from weather stations operated by Met Eireann at Shannon Airport (Met Eireann, 2018).
Appendix E

Supporting Information -
Greenhouse gas emissions from a willow evapotranspiration system
Figure E.1: CO₂ flux values vs water temperature and water level in inspection well, air temperature, atmospheric pressure, evapotranspiration, relative humidity and wind gust. CO₂ fluxes are on log-scale.
Figure E.2: CH$_4$ flux values vs water temperature and water level in inspection well, air temperature, atmospheric pressure, evapotranspiration, relative humidity and wind gust.
Figure E.3: $N_2O$ flux values vs water temperature and water level in inspection well, air temperature, atmospheric pressure, evapotranspiration, relative humidity and wind gust.
Figure E.4: Daily average of CO$_2$ flux values vs daily average of water temperature and water level in inspection well, air temperature, atmospheric pressure, evapotranspiration, relative humidity and wind gust with their mean standard errors.
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