

Sludge-Drying Lagoons: a Potential Significant Methane Source in Wastewater Treatment Plants

- Yuting Pan^{†§}
- Liu Ye^{†‡}
- Ben van den Akker^{||,‡#}
- Ramon Ganigué Pagès^{†∇}
- Ronald S. Musenze[†]
, and
- Zhiguo Yuan[†]

View Author Information

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[†]Advanced Wastewater Management Centre and [‡]School of Chemical Engineering, The University of Queensland, St. Lucia, 4072 Queensland, Australia

[§] Department of Environmental Science and Engineering, School of Architecture and Environment, Sichuan University, Chengdu, Sichuan 610065, China

^{||} Australian Water Quality Centre, Adelaide, 5000 South Australia, Australia

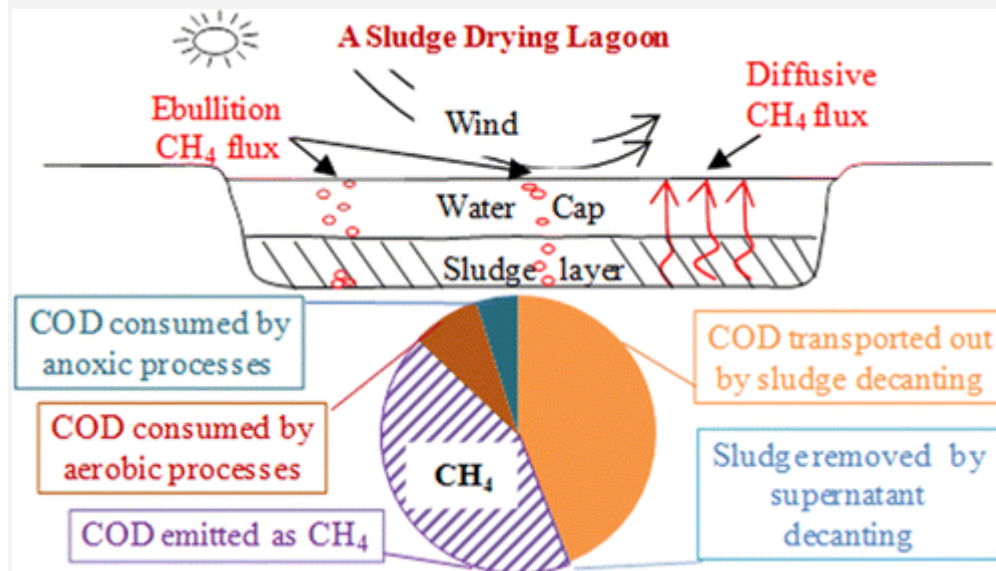
[‡] Health and Environment Group, School of the Environment, Flinders University, Bedford Park, 5042 South Australia, Australia

[#] Centre for Water Management and Reuse, School of Natural and Built Environments, University of South Australia, Mawson Lakes, 5095 South Australia, Australia

[∇] LEQUIA, Institute of the Environment, University of Girona, Girona, Catalonia, 17071 Spain

*Phone + 61-7-3365-4374; fax +61-7-3365-4726; email: zhiguo@awmc.uq.edu.au

Abstract



“Sludge-drying lagoons” are a preferred sludge treatment and drying method in tropical and subtropical areas due to the low construction and operational costs. However, this method may be a potential significant source of methane (CH₄) because some of the organic matter would be microbially metabolized under anaerobic conditions in the lagoon. The quantification of CH₄ emissions from lagoons is difficult due to the expected temporal and spatial variations over a lagoon maturing cycle of several years. Sporadic ebullition of CH₄, which cannot be easily quantified by conventional methods such as floating hoods, is also expected. In this study, a novel method based on mass balances was developed to estimate the CH₄ emissions and was applied to a full-scale sludge-drying lagoon over a three year operational cycle. The results revealed that processes in a sludge-drying lagoon would emit 6.5 kg CO₂-e per megaliter of treated sewage. This would represent a quarter to two-thirds of the overall greenhouse gas (GHG) emissions from wastewater-treatment plants (WWTPs). This work highlights the fact that sludge-drying lagoons are a significant source of CH₄ that adds substantially to the overall GHG footprint of WWTPs despite being recognized as a cheap and energy-efficient means of drying sludge.

1 Introduction

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Excess sludge is an abundant byproduct of WWTPs. The treatment and disposal of this sludge is one of the most challenging and costly components of the wastewater treatment process, which can represent up to 60% of the total treatment cost.⁽¹⁾ Therefore, it is natural for WWTP managers and operators to utilize economical means of sludge treatment that are suited to local conditions. In tropical and subtropical regions, where the climate is hot and dry and land is inexpensive, a “sludge-drying

lagoon” is the preferred sludge treatment and drying method due to its low construction and operating costs.⁽²⁾ A sludge-drying lagoon can be a simple earth basin or a concrete reservoir equipped with some auxiliary works such as a bottom liner to prevent groundwater contamination and a supernatant-decanting facility.⁽³⁾

A typical operational cycle of a sludge-drying lagoon is usually more than one year depending on the local climate and consists of the following major phases: “filling”, “drying”, and “desludging”.⁽⁴⁾ During the filling phase, digested sludge from WWTPs is pumped into the sludge-drying lagoon. When the designed sludge depth is reached, the filling stops, and the drying phase commences. The solids settle and form a thick sludge layer at the base of the lagoon as the sludge is dried by evaporation and is stabilized by microbial processes. Usually, a supernatant water layer of more than 0.5 m is maintained over the digesting sludge layer, forming a “cap”. This allows the oxidation of odorous compounds that are generated and released from the sludge layer during anaerobic digestion.⁽⁵⁾ Once the dewatered sludge reaches the desired solids concentration (e.g., 25% to ~30% dry sludge content), the dried sludge is removed in a single desludging event.

Although sludge-drying lagoons provide a cost-effective and energy-efficient method for treating excess sludge from WWTPs, they may be a significant source of CH₄. This is because part of the organic matter treated in the lagoon would be transformed by microorganisms under anaerobic conditions, leading to CH₄ production. In general, anaerobic digestion is likely to happen at the bottom of the sludge-drying lagoon in the sludge layer where oxygen or other external electron acceptors are depleted (see [Figure 1](#)). In addition to the anaerobic digestion process, organic matter can also be removed by other microbial processes such as aerobic respiration, denitrification, and sulfate reduction. For example, aerobic oxidation of organic matter would occur within the upper “water cap” layer, using oxygen that is continuously diffusing from the atmosphere or produced in situ by algae. This oxygen supply would also drive nitrification, producing nitrate or nitrite, which would facilitate the oxidation of organic matter in anoxic zones via denitrification. Other electron acceptors such as sulfate can also facilitate the oxidation of organic matter to carbon dioxide (CO₂) via sulfate reduction.

Figure 1

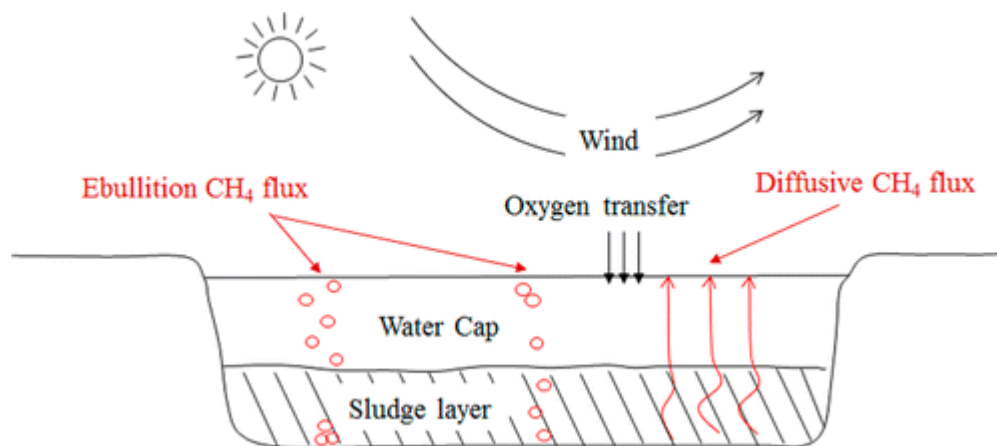


Figure 1. Schematic representation of methane emission from a sludge-drying lagoon.

Understanding CH₄ emissions is of great concern because CH₄ is an important GHG with a global warming potential of around 25 times that of CO₂ equivalents.⁽⁶⁾ However, the CH₄ emissions from full-scale sludge-drying lagoons are rarely being reported due to a number of challenges: (1) a typical sludge-drying lagoon cycle is long (up to several years, depending on the local conditions) and, therefore, emissions are expected to vary over time; (2) significant spatial variation would exist due to sporadic CH₄ and CO₂ gas ebullition⁽⁵⁾ and is unlikely to be captured by the traditional gas-hood method that is widely applied for quantifying emissions from wastewater-treatment plants; and (3) access to sample sludge-drying lagoons is difficult and presents occupational hazards.

The overall aim of this study was two-fold: (1) to develop a method for the quantification of methane emissions from sludge-drying lagoons and (2) to determine the contribution of sludge-drying lagoon treatment processes to the overall carbon footprint of a WWTP. The methodology is demonstrated through its application to a full-scale sludge-drying lagoon in Australia over a 3 year operational cycle.

2 Method Development

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A quantification method based on mass balance was developed to estimate the CH₄ emissions by considering the key physical and biological processes relevant to chemical oxygen demand (COD) balance, total nitrogen (TN) balance, and oxygen balance within a sludge-drying lagoon. The detailed mass balance is schematically shown in [Figure 2](#).

Figure 2

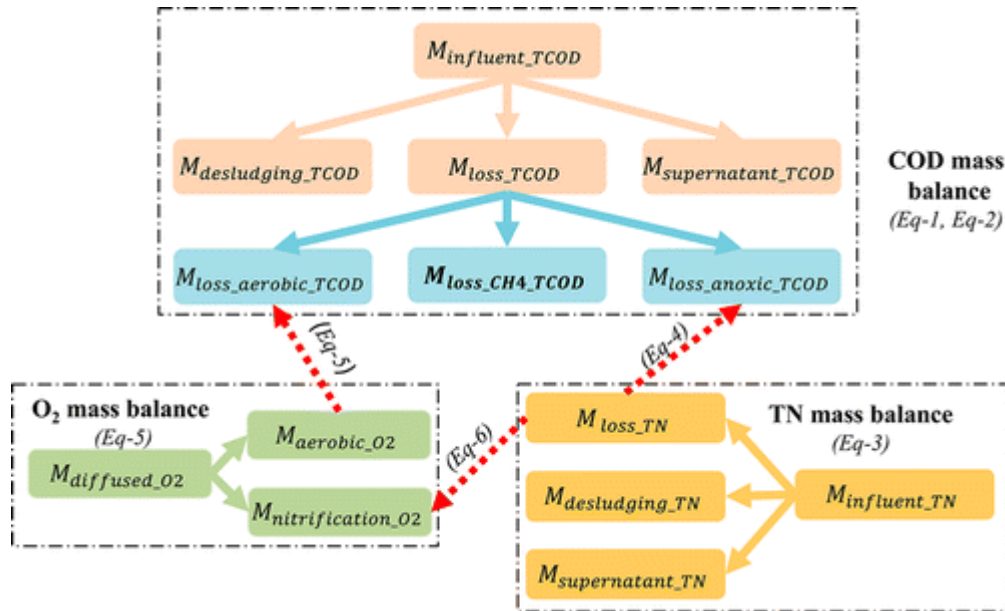


Figure 2. Schematic illustration of the mass balance within a sludge-drying lagoon.

2.1 COD, Nitrogen, and Oxygen Balances of a Sludge-Drying Lagoon

CH₄ Production and COD Mass Balance

As shown in [Figure 2](#), the organic matter contained within the influent sludge ($M_{\text{influent_TCOD}}$) has three possible fates: (1) the COD contained in the remaining dried sludge would be transported out of the sludge lagoon by sludge decanting at the end of the operational cycle ($M_{\text{desludging_TCOD}}$); (2) the COD suspended or dissolved in the supernatant would be removed in the supernatant decanting process ($M_{\text{supernatant_TCOD}}$); and (3) the gap between the COD transported in and the COD transported out will be the “lost” COD ($M_{\text{loss_TCOD}}$). The “lost” COD is due to microbial degradation under aerobic, anoxic, and anaerobic conditions. Under aerobic or anoxic conditions, the COD is lost from the system in the form of CO₂, while under anaerobic conditions, the COD loss is attributed to CH₄ production. Therefore, the amount of CH₄ produced during lagoon treatment can be estimated through eqs [1](#) and [2](#), as shown below:

$$M_{\text{loss_TCOD}} = M_{\text{influent_TCOD}} - M_{\text{desludging_TCOD}} - M_{\text{supernatant_TCOD}} \quad (1)$$

$$M_{\text{loss_CH}_4\text{-TCOD}} = M_{\text{loss_TCOD}} - M_{\text{loss_anoxic_TCOD}} - M_{\text{loss_aerobic_TCOD}} \quad (2) \text{ where:}$$

$M_{\text{loss_TCOD}}$ (tons of TCOD): the total COD loss during one lagoon cycle;

$M_{\text{influent_TCOD}}$ (tons of TCOD): the total COD loaded into the lagoon;

$M_{\text{desludging_TCOD}}$ (tons of TCOD): the total COD removed in desludging;

$M_{\text{supernatant_TCOD}}$ (tons of TCOD): the total COD contained in the supernatant drained;

$M_{\text{loss_CH}_4\text{-TCOD}}$ (tons of TCOD): the amount of CH₄ produced measured as COD;

$M_{\text{loss_anoxic_TCOD}}$ (tons of TCOD): the COD loss through anoxic reactions; and

$M_{\text{loss_aerobic_TCOD}}$ (tons of TCOD): the COD loss through aerobic reactions.

In [eq 1](#), $M_{\text{influent_TCOD}}$, $M_{\text{desludging_TCOD}}$, and $M_{\text{supernatant_TCOD}}$ can be directly calculated based on the lagoon operational data, which was illustrated in this detailed case study. In [eq 2](#), $M_{\text{loss_TCOD}}$ is the output of [eq 1](#), while $M_{\text{loss_anoxic_TCOD}}$ and $M_{\text{loss_aerobic_TCOD}}$ are estimated based on nitrogen balance and oxygen balance, as shown below.

Nitrogen Mass Balance

COD consumption by microorganism within anoxic zones is mainly achieved via denitrification. Therefore, the nitrogen balance is incorporated to estimate COD loss via denitrification, i.e., $M_{\text{loss_anoxic_TCOD}}$. Similar to the COD mass balance, the total nitrogen contained within the influent sludge ($M_{\text{influent_TN}}$) is removed via three possible mechanisms: (1) transported out of the sludge lagoon by the desludging process at the end of a sludge-drying lagoon operational cycle ($M_{\text{desludging_TN}}$); (2) removed from the supernatant decanting process ($M_{\text{supernatant_TN}}$); and (3) the difference between the TN transported in and the TN transported out would be the “lost” TN ($M_{\text{loss_TN}}$) caused by microbial denitrification that transforms solid or dissolved nitrogen into nitrogen gas, mainly N_2 ; N_2O emissions are expected to be negligible, which was confirmed during the sampling campaign. Therefore, the “lost” nitrogen matter through the microbial denitrification processes can be calculated according to [eq 3](#):

$M_{\text{loss_TN}} = M_{\text{influent_TN}} - M_{\text{desludging_TN}} - M_{\text{supernatant_TN}}$ (3) where:

$M_{\text{loss_TN}}$ (tons of TN): the total nitrogen loss during the lagoon operation;

$M_{\text{influent_TN}}$ (tons of TN): the total amount of nitrogen matter loaded into the lagoon;

$M_{\text{desludging_TN}}$ (tons of TN): the nitrogen matter removed in desludging; and

$M_{\text{supernatant_TN}}$ (tons of TN): the nitrogen matter contained in the supernatant drained.

The stoichiometric COD consumption for denitrification is 2.86 g of COD/g N. Assuming nitrate is the electron acceptor, the COD loss through anoxic reactions can be calculated as $M_{\text{loss_anoxic_TCOD}} = 2.86 \times M_{\text{loss_TN}}$ (4) Nitrite could be an alternative intermediate form of oxidized nitrogen that occurs during nitrification and denitrification. In this case, the reaction stoichiometry would be 1.71 g of COD/g N. Therefore, the true ratio would be between 1.71 and 2.86 g of COD/g N, depending on the contribution of each intermediate. However, as shown in the oxygen balance below, the uncertainty associated with the COD/N ratio does not affect the estimated amount of CH_4 emitted.

Oxygen Mass Balance

The oxygen balance was also introduced to estimate the COD loss due to aerobic respiration, i.e., $M_{\text{loss_aerobic_TCOD}}$. Oxygen for aerobic oxidation of organic matter and nitrification will be continuously diffused from the atmosphere into the liquid phase due to air circulation. As discussed below, oxygen production from phototrophic processes

does not affect the COD balance and, therefore, was not included. The mass of oxygen transferred during the operational life of a lagoon can be estimated basing on water–air gas-transfer models (further described in [section 2.2](#)). Once the quantity of oxygen diffused into the liquid ($M_{\text{diffused_O}_2}$, tons of O_2) and the oxygen consumed by nitrification ($M_{\text{nitrification_O}_2}$, tons of O_2) were quantified, the aerobic heterotrophic respiration was estimated using [eq 5](#): $M_{\text{loss_aerobic_TCOD}} = M_{\text{aerobic_O}_2} = M_{\text{diffused_O}_2} - M_{\text{nitrification_O}_2}$ (5) where:

$M_{\text{aerobic_O}_2}$ (tons of oxygen): the amount of oxygen consumed through the aerobic COD oxidation process;

$M_{\text{diffused_O}_2}$ (tons of oxygen): the total amount of oxygen diffused from the atmosphere into the sludge-drying lagoon; and

$M_{\text{nitrification_O}_2}$ (tons of oxygen): the amount of oxygen consumed for nitrification;

The oxidized nitrogen transformed by denitrification is mainly provided by nitrification. Assuming that ammonium is oxidized all the way through to nitrate and then denitrified to nitrogen gas, the amount of oxygen consumed for nitrification is $4.57 \text{ g O}_2/\text{g N}^7$. The amount of oxygen used by nitrification can then be estimated using [eq 6](#):

$M_{\text{nitrification_O}_2} = 4.57 \times M_{\text{loss_TN}}$ (6) If nitrite were the end product of nitrification, the oxygen consumption coefficient would be $3.43 \text{ g of O}_2/\text{g of N}$. Therefore, the true oxygen consumption for nitrification would be between 3.43 and $4.57 \text{ g of O}_2/\text{g of N}$.

Introducing [eqs 4](#), [5](#), and [6](#) into [eq 2](#), we have,

$M_{\text{loss_CH}_4\text{-TCOD}} = M_{\text{loss_TCOD}} - M_{\text{diffused_O}_2} + 1.71 \times M_{\text{loss_TN}}$ (7) The coefficient $1.71 \text{ g of COD/g of N}$ in [eq 7](#) is independent of the intermediate (nitrate or nitrite) between nitrification and denitrification.

In the above balance analysis, we have ignored COD oxidation by sulfate and also the potential for oxygen formation by algal photosynthesis because these processes are not expected to affect the overall estimation of $M_{\text{loss_CH}_4\text{-TCOD}}$. While consuming COD, sulfate reduction forms hydrogen sulfide. The lagoon is capped by water to avoid hydrogen sulfide emission. This water layer leads to the oxidation of sulfide to sulfate, and thus, oxygen is consumed. The amount of oxygen consumed for sulfide oxidation is equivalent to the amount of COD consumed for sulfate reduction, and, therefore, the overall COD and oxygen balance is not affected. Similarly, any oxygen produced by algae is accompanied by CO_2 fixation to organic carbon. Therefore, algal photosynthesis does not affect the overall COD and oxygen balances.

Another reaction we have ignored is the anammox reaction, which could happen at the interface of the aerobic and anaerobic zones in the sediments. In this zone, nitrite formed by ammonium oxidation could react with ammonium to produce N_2 . The combined nitrification and anammox reaction is $2\text{NH}_3 + \frac{3}{2}\text{O}_2 \rightarrow \text{N}_2 + 3\text{H}_2\text{O}$. This means that the net oxygen consumption for NH_3 conversion to N_2 is still $1.71 \times M_{\text{loss_TN}}$. In other words, the presence of the anammox reaction does not affect [eq 7](#). In the above combined reaction, for simplicity, we have ignored nitrate production from nitrite by the anammox reaction. As discussed above, denitrification from nitrite or nitrate does not affect the oxygen balance, either.

2.2 Estimation of Oxygen Diffusion Using Wind Models

The amount of oxygen transferred ($M_{\text{diffused}_O_2}$) during one operational cycle of a sludge-drying lagoon can be quantified based on [eq 8](#): $M_{\text{diffused}_O_2} = \int_0^T F(t) \times A \times dt$ (8) where $F(t)$ ($\text{g/m}^2/\text{day}$) is the oxygen-transfer flux from air to water; A (m^2) is the area of the sludge-drying lagoon; T (d) is the duration of one operational cycle of a sludge-drying lagoon. For the estimation of F , we used the thin-boundary-layer equation that is detailed below.

Estimation of Oxygen-Transfer Flux Using the Thin-Boundary-Layer Equation

$F = k(C_{\text{obs}} - C_{\text{sat}})$ (9) where F is the estimation of oxygen-transfer flux, k (m/d) is the normalized gas-transfer velocity, C_{obs} (g/m^3) is surface water dissolved oxygen concentration, and C_{sat} (g/m^3) is the atmospheric equilibrium and saturation concentration at the prevailing in situ temperature. C_{obs} can be measured (as detailed in the case study), while C_{sat} can be calculated using Henry's law from the atmospheric concentration of oxygen. For the estimation of k , we used wind-based models as explained below.

Estimation of the Gas-Transfer Velocity

Although many factors such as wind, waves, bubbles, and water currents can influence gas exchange across the water–air interface, [\(8\)](#) wind is clearly recognized as being key for stationary water bodies such as the water layer in a sludge-drying lagoon. [\(9, 10\)](#) To estimate the oxygen-transfer velocity due to wind (k), we used three commonly used wind models, including Ro and Hunt (2006) [\(9\)](#) (hereafter RH06), Liss and Merlivat (1986) [\(11\)](#) (hereafter LM 86), and Cole and Caraco (1998) [\(10\)](#) (hereafter CC98) (see the [Supporting Information](#) for details). RH06 is a unified model for wind-driven surficial oxygen transfer into stationary water bodies developed by combining coefficients from relationships for gas transfers derived in investigations in controlled wind tunnels, floating reaerators in open water, and natural open-water bodies. LM86 was developed through laboratory experimental approaches using wind and water tunnels and validated with measurements from natural open water bodies, while CC98 was developed using wind measurements and gas fluxes in freshwater lakes. Details regarding these models including variability in their estimates have been previously discussed. [\(12-14\)](#)

3 Implementing the Method Developed to a Full-Scale Sludge-Drying Lagoon

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3.1 Sludge-Drying Lagoon Used for the Case Study

The sludge-drying lagoon treatment process used in this case study receives anaerobically digested sludge from two Australian WWTPs that have design capacities of 160 ML/day and 45 ML/day. The sludge is composed of both digested primary and digested secondary (waste-activated) sludge. The climate in the area is classified as subtropical, with hot, dry summers and mild winters with moderate rainfall. There are eight evaporation sludge-drying lagoons, with a total surface area of 120 ha and a design depth of 1.5 m. Lagoon filling is spread across several lagoons until one is full. The filled lagoon is moved offline, and a new lagoon replaces its rotation. Sludge in the offline lagoon then undergoes drying and stabilization before desludging at the end of the operational cycle.

We investigated one of the eight lagoons. The lagoon operation involves a three-year operational cycle, including a 27 month intermittent filling, eight months of drying and one month of desludging. The initial “water cap” depth was 1 m; however, the volume could change due to evaporation and rainfall. Excess supernatant was collected and returned to the head of the plant. Sludge removed from the lagoons is stockpiled for a three year period to be reused on farmland or trucked offsite for landfill disposal.

3.2 Data Collection

Operation Regime and the Parameters of the Sludge-Drying Lagoon

The operation information on a sludge-drying lagoon was collected from the operator, which included the following information that was needed to calculate the TCOD mass and TN mass required by eqs [1](#) and [3](#):

- (a) *During the sludge “feeding period”*: parameters relevant to the TCOD mass and TN mass fed into the sludge-drying lagoon collected included: (1) the monthly sludge influent flow rate (Q_{inf} , ML/month); (2) the dry sludge (total suspended solid) content in the influent (S_{inf_TS} , percent of dry sludge weight/the total influent weight); (3) the influent TCOD concentration (S_{inf_TCOD} , kg of COD/kg of dry sludge); and (4) the influent TN concentration (S_{inf_TN} , kg of TKN/kg of dry sludge).
- (b) *During the “desludging period”*: parameters relevant to the TCOD mass and TN mass removed from the sludge-drying lagoon collected included: (1) the weight of the sludge removed in the desludging process (M_{des} , tons); (2) the dry sludge (total suspended solids) content in the removed sludge (S_{des_TS} , percent of dry sludge weight/ M_{des}); (3) the TCOD concentration in the removed sludge (S_{des_TCOD} , mg of TCOD/kg of dry sludge weight); and (4) the TN concentration in the removed

sludge ($S_{\text{des_TN}}$, mg of TN/kg of dry sludge weight).

(c)

Sludge supernatant parameters: parameters relevant to the TCOD mass and TN mass content in the supernatant collected included: (1) volume of the supernatant (V_{sup} , ML); (2) the supernatant TCOD concentration (mg of TCOD/L); and (3) the supernatant TN concentration ($S_{\text{sup_TN}}$, mg of TKN/L).

Wind Data Collection

A total of three different wind models, which all require wind-speed data, were used to estimate the oxygen diffusion. The wind speed data, spanning around two years, was obtained from two Bureau of Meteorology automatic weather and climate stations located closest to the lagoon. Wind data was measured every 10 s and the averages logged at 15 min intervals throughout the study period. The 15 min averages were then used for the estimation of k using the three models, (15) which was subsequently used for the estimation of F and $M_{\text{diffused_O}^2}$ using eqs 9 and 8, respectively.

Determination of the Surface Water Dissolved Oxygen Concentration C_{obs}

A remote-controlled boat (Figure S1) equipped with a DO meter (YSI Professional Plus) was used to monitor the surface DO concentrations. The remote controlled boat could travel approximately 50 m away from the bank of the sludge-drying lagoon to record DO measurement at different locations. The DO meter was mounted at the front of the boat and programmed to record the DO concentration and temperature at 5 min intervals. The DO sensor was submerged ~2 cm below the sludge-drying lagoon water surface.

4 Results

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4.1 Operational Regime and Parameters of the Sludge-Drying Lagoon

The parameters used to calculate the mass of TCOD and TN that was (i) fed into the sludge-drying lagoon, (ii) removed from sludge-drying lagoon during the desludging period, and (iii) removed when decanting the supernatant are summarized in Table 1, which shows parameter range and average and standard error. Average values were used to calculate the mass of TCOD and TN transported in and out of the sludge-drying lagoon. To calculate the overall volume of sludge fed into the sludge-drying lagoon during the sludge “feeding period”, we integrated the monthly influent flow-rate data

(Q_{inf}). The volume of supernatant removed from the sludge-drying lagoon was not collated because routine decanting does not occur during normal operation. Occasional ad hoc decanting of supernatant does occur; however, the flow was not recorded by the plant operator. We assumed that 15 ML was removed from the plant because the surface area of the lagoon studied is 15 ha and the initial water was 1 m. The potential impact of this uncertainty associated with the volume of supernatant removed was shown to be negligible, as detailed in [section 4.3](#).

Table 1. Parameters Relevant to TCOD and TN Mass-Balance Analysis in a Complete Operational Cycle of the Sludge-Drying Lagoon Studied

parameter	definition
sludge-drying lagoon influent parameters	
Q_{inf}	monthly influent flow rate (ML/month)
S_{inf_TS}	dry sludge (total suspend solid) content in the influent (percent of dry sludge weight/the total i
S_{inf_TCOD}	influent TCOD concentration (kg of TCOD/kg of dry sludge)
S_{inf_TKN}	influent TKN concentration (kg of TKN/kg of dry sludge)
$S_{inf_NO_3^-}$	influent NO_3^- concentration (mg NO_3^- -N/L)
$S_{inf_NO_2^-}$	influent NO_2^- concentration (mg NO_2^- -N/L)
parameters collected during lagoon desludging process	
M_{des}	weight of the dry sludge removed in the desludging process (kilotons)
S_{des_TS}	dry sludge (total suspend solid) content in the removed sludge (% of dry sludge weight/ M_{des})
S_{des_TCOD}	TCOD concentration in the removed sludge (mg TCOD/kg dry sludge weight)

parameters collected during lagoon desludging process

$S_{\text{des_TKN}}$ TKN concentration in the removed sludge (mg TKN/kg dry sludge weight)

sludge-drying lagoon supernatant parameters

V_{sup}	volume of supernatant (ML)	200
$S_{\text{sup_TCOD}}$	supernatant TCOD concentration (mg TCOD/L)	134.1
$S_{\text{sup_TKN}}$	supernatant TN concentration (mg TKN/L)	18.1
$S_{\text{sup_NO}_3^-}$	supernatant NO_3^- concentration (mg NO_3^- -N/L)	0.41
$S_{\text{sup_NO}_2^-}$	supernatant NO_2^- concentration (mg NO_2^- -N/L)	0

a

Plant A sludge.

b

Plant B sludge.

The dissolved CH_4 and N_2O concentrations in the surface water were measured in two sludge-drying lagoons, with Lagoon A representing a maturing lagoon while Lagoon B represented a matured lagoon (Figure S2). The CH_4 concentration varied significantly, with concentrations of 0.02 mg of CH_4 /L and 0.9 mg of CH_4 /L, respectively. Such variation was probably closely related to the level of maturity of a sludge-drying lagoon. In contrast, no large difference was observed in N_2O concentrations, which were, on average, 0.025 mg N/L within both lagoons. These concentrations were measured to confirm the presence of CH_4 and N_2O in the lagoon and were not required to calculate emissions.

4.2 Estimation of the Amount of Oxygen Transferred

The measured DO concentration of the lagoon surface water was 3.7 ± 0.3 mg/L, with no pronounced spatial or temporal variations. This was probably because the lagoon is shallow, and the sludge is evenly distributed at the bottom of the lagoon. A total of three wind-based models were used to estimate the gas-transfer velocities and eventually fluxes, with the estimated yearly average oxygen-transfer fluxes (F) shown in Figure 3.

Figure 3

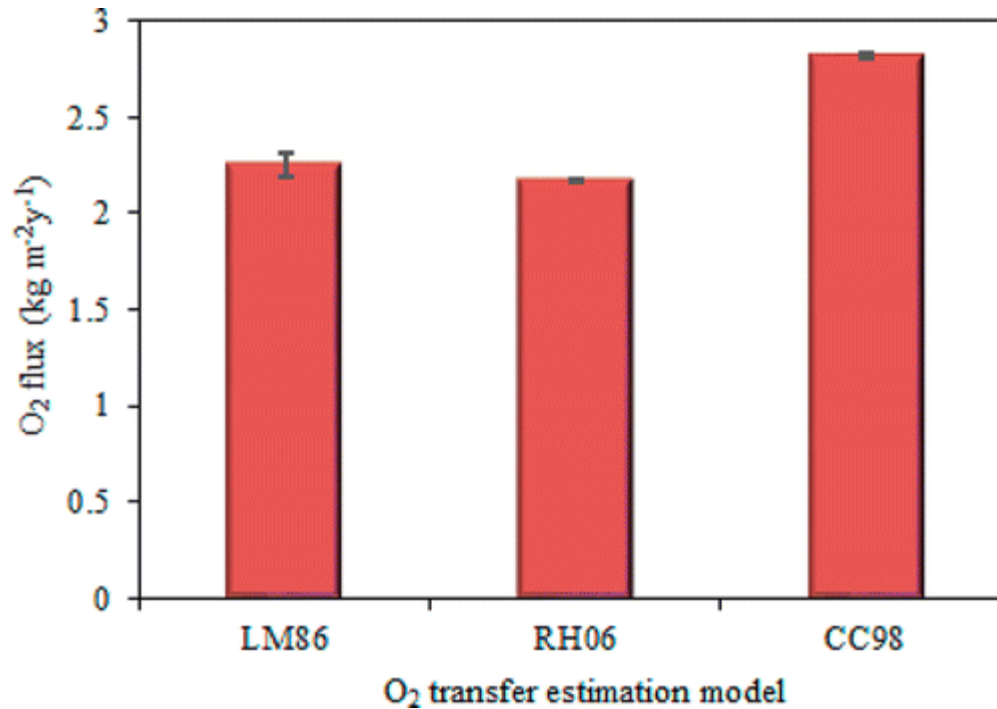


Figure 3. Estimated O₂-transfer flux based on different wind models (LM86, Liss and Merlivat (1986);(11) RH06, Ro and Hunt (2006);(9) CC98, Cole and Caraco (1998)).(10)

The estimated F varied between the three models, ranging between 2.2 to 2.8 kg O₂/(m² × year). The average F value (2.4 ± 0.35 kg O₂/(m² × year)) that was derived from the three models was applied to [eq 7](#), together with (1) the surface area of 15 hectare and (2) the sludge filling and drying period of 35 months. The oxygen transfer from air to sludge-drying lagoon ($M_{\text{diffused}_O_2}$) was estimated to be 1.1 ± 0.35 kilotons of O₂.

4.3 Mass Balance of the Sludge-Drying Lagoon Investigated and CH₄ Emission

With the plant operation data acquired ([Table 1](#)), the component required by the TCOD and TN balance in [eq 1](#) and [eq 3](#) can be calculated, with the results shown in [Table 2](#). For example, $M_{\text{influent_TCOD}}$ is the sum of the TCOD origin from Plant A and Plant B. Take Plant A as an example, the COD input was calculated by multiplying the overall volume of sludge fed into the sludge-drying lagoon during the sludge “feeding period” (Q_{inf}), the density of the influent sludge (ρ), the dry sludge content in the influent ($S_{\text{inf_TS}}$), and the influent TCOD concentration ($S_{\text{inf_TCOD}}$). As shown in [Table 1](#), the $S_{\text{inf_TS}}$ and the $S_{\text{inf_TCOD}}$ for Plant A were 2.0 ± 0.022% and 0.87 ± 0.025 kg of TCOD/kg of dry sludge, respectively. The density of the sludge was assumed to be equal to the density of water. Therefore, the COD input from Plant A was estimated to be 3.9 kilotons of TCOD. Following the same method, the COD input for Plant B was estimated to be 2.8 kilotons of TCOD. $M_{\text{influent_TCOD}}$ is the sum of the TCOD origin from Plant A and Plant B, which was 6.7 kilotons of TCOD, as shown in [Table 2](#). Similarly, the amount of TCOD and TN removed during the desludging process ($M_{\text{desludging_TN}}$), and supernatant decanting

($M_{\text{supernatant_TCOD}}$, $M_{\text{supernatant_TN}}$) was calculated based on parameters listed in [Table 1](#), with the results shown in [Table 2](#). The difference between the COD/TN transported in and the COD/TN transported out represents the “lost” COD ($M_{\text{loss_TCOD}}$)/TN ($M_{\text{loss_TN}}$), which was determined using eqs [1](#) and [3](#), respectively.

Table 2. COD, TN, and Oxygen Balance of the Sludge-Drying Lagoon Studied

	COD mass balance (Eq-1) (kilo tonnes TCOD)		TN mass balance (Eq-3) (kilo tonnes TN)		Oxygen mass balance (Eq-5) (kilotonnes O ₂)	
	$M_{\text{influent_TCOD}}$	6.7 ± 0.08	$M_{\text{influent_TN}}$	0.59 ± 0.01	$M_{\text{diffused_O2}}$	1.1 ± 0.11
	$M_{\text{desludging_TCOD}}$	2.9 ± 0.18	$M_{\text{desludging_TN}}$	0.47 ± 0.02	$M_{\text{nitrification_O2}}$	0.52 ± 0.09
	$M_{\text{supernatant_TCOD}}$	negligible	$M_{\text{supernatant_TN}}$	negligible	$M_{\text{aerobic_O2}}$	0.53 ± 0.14
	$M_{\text{loss_TCOD}}$	3.8 ± 0.20	$M_{\text{loss_TN}}$	0.11 ± 0.02		
		↓		↓		↓
		(Eq-4)		(Eq-5)		
COD mass balance (Eq-2) (tonnes TCOD)	$M_{\text{loss_TCOD}}$	3.8 ± 0.28	$M_{\text{loss_anoxic_TCOD}}$	0.33 ± 0.06	$M_{\text{loss_aerobic_TCOD}}$	0.53 ± 0.14
	$M_{\text{loss_CH4_TCOD}}$	2.86 ± 0.32				

Once $M_{\text{loss_TN}}$ was determined, the amount of oxygen consumed for nitrification $M_{\text{nitrification_O}_2}$ was calculated using [eq 6](#), and the COD lost through anoxic reactions ($M_{\text{loss_anoxic_TCOD}}$) was calculated using [eq 4](#). The amount of oxygen used by heterotrophs aerobically $M_{\text{aerobic_O}_2}$ can be estimated by the oxygen balance ([eq 5](#)).

Finally, using [eq 2](#), we estimated the mass of CH₄ generated in the sludge-drying lagoon to be 2.86 ± 0.32 kilotons of COD over a period of 3 years. Considering that 1 g of CH₄ is equivalent to 4 g of COD, and the global warming effect of 1 g of CH₄ correspond to 25 g of CO₂ equivalents with a time horizon of 100 years, the CH₄ emission from the sludge-drying lagoon investigated was 6.1 ± 0.67 kilotons of CO₂-e/year. There are eight similar sludge-treatment lagoons operated in parallel; therefore, the overall CH₄ emission from the sludge-treatment lagoon system was estimated to be 48.9 ± 5.5 kilotons of CO₂-e/year.

With the influent COD considered as 100% bioavailable COD, [Figure 4](#) shows that 32% of the influent COD was removed by the desludging process, whereas <1% of COD was removed by the supernatant decanting, while the remainder of the COD was “lost” due to microbial activities. The aerobic, anoxic, and anaerobic processes all contribute significantly to COD elimination, which accounted for 8 ± 0.9%, 5 ± 0.3%, and 43 ± 5%, respectively, of the influent COD.

Figure 4

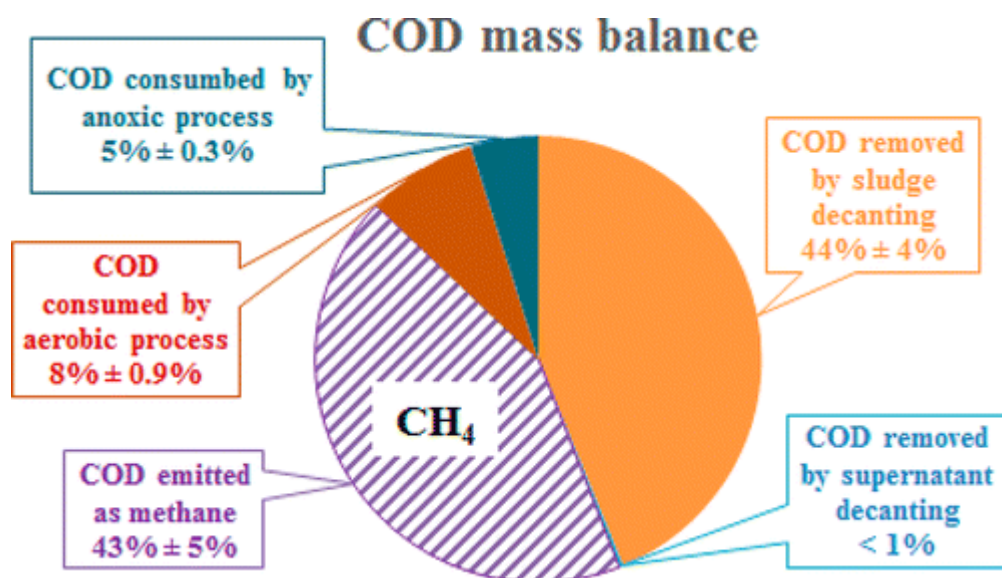


Figure 4. COD mass balance for the sludge-drying lagoon studied.

5 Discussion

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5.1 Estimation of Methane Emissions from Sludge-Drying Lagoon

With the increasing awareness of greenhouse gas (GHG) emission of WWTPs, the monitoring of many full-scale plants has been carried out in recent years. Currently, the most widely adopted quantification method is through the use of online gas hoods to capture emitted gases.[\(16, 17\)](#) This method has been shown to provide reliable results for aeration tanks with more or less evenly distributed aerators.[\(18\)](#) However, gas hood measurements are not a suitable method to quantify CH₄ from sludge-drying lagoon systems. This is partially because the emission is expected to have significant temporal variability during operation cycle, as shown in [Figure S2a](#) that effective quantification of the emissions would require a sampling campaign to last for the entire operation cycle. More importantly, the random gas ebullition cannot be properly captured by a limited number of gas hoods. To date, there is no information on the contributions of bubbling and diffusive CH₄ fluxes to the overall CH₄ emission from a sludge-drying lagoon. However, research from other stationary water systems, such as lakes, has found that the bubble form of CH₄ significantly contributes to CH₄ emissions.[\(19, 20\)](#)

In this work, a novel method was developed that integrated TCOD, TN, and oxygen balances to estimate CH₄ emissions from sludge-drying lagoon systems. The method only requires some routine operational data from a sludge-drying lagoon, such as concentrations of TCOD and TN and flow measurements, which are detailed in [Table 1](#). Additional data related to oxygen transfer, such as the wind speed and surface oxygen concentration in the lagoon, can be relatively easily acquired, as was demonstrated in this case study.

It should be noted that the CH₄ emissions value estimated using the methodology developed here was not validated with an independent method simply due to the lack of independent methods available. Although further verification of the method is required, the method is promising and will likely provide a simple and useful tool to estimate CH₄ emissions from sludge-drying lagoons.

It is also worthwhile to mention that sludge-drying lagoons also likely emit N₂O, another potent greenhouse gas. Indeed, the surface-water N₂O concentration we measured was approximately 0.024 mg N₂O/L, which is over 70 times oversaturated (the saturation N₂O level is approximately 3.3×10^{-4} mg N₂O/L, assuming an atmospheric N₂O mixing ratio of 325 ppb). The results indicate that the sludge-drying lagoon studied is indeed a source of N₂O. The proposed methodology, however, is not directly applicable to the estimation of N₂O emissions.

5.2 Significance of Methane Emission from Sludge-Drying Lagoons to the Overall Carbon Footprint of a WWTP

The overall GHG emissions from WWTPs are associated with the on-site emissions and off-site emissions. On-site GHG emissions are related to the wastewater and sludge treatment. Off-site emissions are related to the power generation for plant operation, production, and transportation of chemicals for on-site usage and degradation of remaining constituents in the effluent, which may result in CH₄ and N₂O emissions, as well as the transportation and disposal of solids. [\(21\)](#) The energy recovered through the use of biogas produced by a plant can offset some of the emissions.

For a WWTP adopting biological nitrogen removal (BNR) processes to remove COD and nitrogen, and sludge digestion processes to treat excess sludge with produced biogas used for energy production, Bani Shahabadi et al. [\(21\)](#) estimated that the overall carbon footprint of a plant, including both the onsite and offsite GHG emissions, was 2694 kg CO₂-e/(ML of sewage treated). De Haas et al. [\(22\)](#) collected operational data from thirty-five wastewater treatment plants in South-East Queensland and estimated that the overall GHG emissions from the plants varied between 1000 kg CO₂-e per ML of sewage treated to 2500 kg CO₂-e per ML of sewage treated. In this study, the sludge inflow originated from two plants with BNR and sludge digestion processes, with a total treatment capacity of 205 ML/day. Considering that the overall CH₄ emission from the sludge-drying lagoons is 48.9 kilotons of CO₂-e/year, the sludge-drying lagoon process would emit 654 kg of CO₂-e/(ML sewage treated). Assuming that the emissions from these plants fall in the range reported so far, this would account for 24–65% of the overall GHG emissions from the two WWTPs generating sludge to the sludge-drying lagoons,

The outcome of this study informs WWTP designers that despite sludge treatment in sludge-drying lagoons being of a relatively low cost, it may considerably increase the overall GHG emissions from a WWTP. Subjective to more field studies, alternative treatment processes may need to be considered to replace sludge-drying lagoons that are being commonly used in tropical and subtropical regions.

Supporting Information

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The Supporting Information is available free of charge on the [ACS Publications website](#) at DOI: [10.1021/acs.est.5b04844](https://doi.org/10.1021/acs.est.5b04844).

- Figures showing the remotely controlled boat used to monitor the lagoon surface DO concentration and the surface water CH₄ and N₂O concentrations in two sludge-drying lagoons. Models used for estimating the gas-transfer velocity. ([PDF](#))

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Author Information

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-
- **Corresponding Author**
 - **Zhiguo Yuan** - †*Advanced Wastewater Management Centre and ‡School of Chemical Engineering, The University of Queensland, St. Lucia, 4072 Queensland, Australia;* Department of Environmental Science and Engineering, School of Architecture and Environment, Sichuan University, Chengdu, Sichuan 610065, China; Australian Water Quality Centre, Adelaide, 5000 South Australia, Australia; Health and Environment Group, School of the Environment, Flinders University, Bedford Park, 5042 South

Australia, Australia; Centre for Water Management and Reuse, School of Natural and Built Environments, University of South Australia, Mawson Lakes, 5095 South Australia, Australia; LEQUIA, Institute of the Environment, University of Girona, Girona, Catalonia, 17071 Spain; Email: zhiguo@awmc.uq.edu.au

- **Authors**

- **Yuting Pan** - Department of Environmental Science and Engineering, School of Architecture and Environment, Sichuan University, Chengdu, Sichuan 610065, China
- **Liu Ye** - †Advanced Wastewater Management Centre and ‡School of Chemical Engineering, The University of Queensland, St. Lucia, 4072 Queensland, Australia; Department of Environmental Science and Engineering, School of Architecture and Environment, Sichuan University, Chengdu, Sichuan 610065, China; Australian Water Quality Centre, Adelaide, 5000 South Australia, Australia; Health and Environment Group, School of the Environment, Flinders University, Bedford Park, 5042 South Australia, Australia; Centre for Water Management and Reuse, School of Natural and Built Environments, University of South Australia, Mawson Lakes, 5095 South Australia, Australia; LEQUIA, Institute of the Environment, University of Girona, Girona, Catalonia, 17071 Spain
- **Ben van den Akker** - Australian Water Quality Centre, Adelaide, 5000 South Australia, Australia; Health and Environment Group, School of the Environment, Flinders University, Bedford Park, 5042 South Australia, Australia; Centre for Water Management and Reuse, School of Natural and Built Environments, University of South Australia, Mawson Lakes, 5095 South Australia, Australia
- **Ramon Ganigué Pagès** - LEQUIA, Institute of the Environment, University of Girona, Girona, Catalonia, 17071 Spain
- **Ronald S. Musenze** - †Advanced Wastewater Management Centre and ‡School of Chemical Engineering, The University of Queensland, St. Lucia, 4072 Queensland, Australia; Department of Environmental Science and Engineering, School of Architecture and Environment, Sichuan University, Chengdu, Sichuan 610065, China; Australian Water Quality Centre, Adelaide, 5000 South Australia, Australia; Health and Environment Group, School of the Environment, Flinders University, Bedford Park, 5042 South Australia, Australia; Centre for Water Management and Reuse, School of Natural and Built Environments, University of South Australia, Mawson Lakes, 5095 South Australia, Australia; LEQUIA, Institute of the Environment, University of Girona, Girona, Catalonia, 17071 Spain

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- **Notes**

The authors declare no competing financial interest.

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