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Long-term onsite monitoring of a sewage sludge drying pan finds methane emissions consistent with IPCC default emission factor

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ABSTRACT

As the wastewater sector moves towards achieving net zero greenhouse gas (GHG) emissions, quantifying and understanding fugitive emissions from various sewage treatment steps is crucial for developing effective GHG abatement strategies. Methane (CH_4) emissions from a sludge drying pan (SDP) were measured at a wastewater treatment plant in Australia for more than a year, using a micrometeorological technique paired with open-path lasers. The emission rate was tightly associated with sludge additions, climatology, and operational processes. The mean emission rate during the 90 weeks after initial sludge addition was 2.3 ($\pm$ 0.8) g m^{-2} d^{-1}, with cumulative emissions of approximately 32 t of CH_4. A dynamic temporal pattern of emissions was observed, highlighting the importance of continuous (or near-continuous) measurements for quantifying SDP emissions. A Methane Correction Factor (MCF) expressed as a fraction of the measured chemical oxygen demand of the sludge, was determined to be 0.17 after 63 weeks (the median operational cycle duration at the facility). This is broadly consistent with, albeit slightly less than, the IPCC default value of 0.2 for shallow anaerobic lagoons. These emission measurements will support wastewater utilities that employ open air sludge drying processes to develop effective GHG abatement strategies.

Introduction

Wastewater treatment facilities are a significant source of greenhouse gas (GHG) emissions to the atmosphere, including methane (CH_4) from the anaerobic treatment of organics, and nitrous oxide (N_2O) from the biological removal of nitrogen (N) through nitrification and denitrification processes (Czepiel et al. 1993, IPCC 2006). Global population rise has increased the need for wastewater treatment. Without emission mitigation efforts, this will lead to increasing GHG emissions from the wastewater sector. It was reported that emissions from waste treatment plants contributed approximately 1% of national GHG emissions in Greece (Koutsou et al. 2018), 2.2% of U.S. CH_4 emission (USEPA 2023), and 3.6% of national emissions in China (Zhao et al. 2023). Bogner et al. (2007) reported that global wastewater CH_4 emissions have increased by 49% from 1990 to 2020, with most of the increase attributed to developing countries, especially in south and east Asia (58% vs 65%), and smaller increases from European countries (4% vs 3%). There is growing interest in reducing GHG emissions from water treatment plants worldwide, and the magnitude of GHG emissions is becoming an important factor in assessing the performance of water treatment plants (Mohsenpour et al. 2021).

Also known as sludge drying beds or sludge drying lagoons, open air sludge drying in sludge drying pans (SDPs) is a widely used method of municipal wastewater sludge dewatering where land availability and climate allow. In particular it is common throughout the United States, Russia, Eastern Europe, and Africa (Elbaz et al. 2020). Open air sludge drying is potentially a significant GHG source in wastewater treatment plants (WWTPs) (Daelman et al. 2012). During sludge drying, CH_4 is produced as the sludge entering the drying pan still has considerable residual CH_4 potential (Daelman et al. 2012). In one particular case it was estimated, based on a mass-balance analysis, that an SDP receiving anaerobically digested sludge accounted for 25–65% of the overall GHG emissions from a WWTP (Pan et al. 2016). However, direct measurement of emissions from SDPs are rarely reported because of the challenges associated with full-scale monitoring, such as the long operation cycles (up to several years), significant spatial variation, and

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occupational hazards to access the SDPs.

In the absence of direct monitoring, wastewater facilities are estimating CH_4 emissions using the default IPCC emission factor. In this approach, emissions are related to the input and output levels of the chemical oxygen demand (or biological oxygen demand) in the sludge. However, there is limited evidence validating the application of this emission factor to this particular process (Czepiel et al. 1993, Moore et al. 2023). There is thus a need for direct measurements at SDPs to validate and refine the emission factor.

The direct measurement of emissions from large open sources like SDPs is challenging (Tremblay et al. 2005). In many jurisdictions the flux chamber technique is the measurement standard. However, chamber measurements are problematic. Chambers modify the environmental conditions inside the chamber, which can potentially alter the ambient emission rate. Further, the small size of the chamber means the measurement may be unrepresentative of the source as a whole, necessitating a substantial measurement survey for large sources (Delre et al. 2017). World-wide efforts are underway to implement measurement methodologies that are more accurate, cheaper, and logistically simpler to use, such as plant-scale tracer methods (Delre et al. 2017, Yoshida et al. 2014, Yver Kwok et al. 2015). One alternative is inverse-dispersion modelling (IDM), a micrometeorological technique where emission rates are calculated from gas concentration measured in the air downwind of the source. The IDM technique measures emissions over a much larger area than chambers, it does not alter the source environment, it has modest measurement requirements, and is well-suited for long-term measurements. IDM has been widely used to quantify emission sources, such as gas wells (Riddick et al. 2019), tailings ponds (You et al. 2021), biogas plants (Groth et al. 2015), and cattle feedlots (Bai et al. 2015), among other applications.

In this study, we employ the IDM technique to quantify CH_4 emissions from an SDP at a large wastewater treatment plant in Australia. By monitoring CH_4 emissions from an SDP near-continuously over an operation cycle, including periods of filling/decanting, and turning, this study aimed to (1) verify the IPCC default CH_4 emission factor for SDPs (Bartram et al. 2019); (2) reveal the temporal dynamics of CH_4 emissions from SDPs, including the contribution from each operational stage; (3) develop a simple mathematical model to estimate CH_4 emissions from SDPs and identify key parameters influencing the emissions, and (4) identify mitigation opportunities.

Results and Discussion

Long-term CH_4 emission rates

The SDP measurements give a near-continuous sequence of emission rates, each a 15-min average, with 6,775 measurements over a period of 613 days. Average weekly emission rates are calculated from this data sequence. Week 1 follows the initial placement of sludge in the SDP on 12–13 March 2021, and measurements began on Week 3, 24 March 2021. The weekly CH_4 emission rates are shown in Fig. 1A, and the weekly precipitation and air temperature are shown in Fig. 1B. Emissions were relatively low during the period of initial sludge feeds (Weeks 1 to 27). During this initial period there was a short-term pattern where emissions rose (and then fell) after each fresh sludge addition. After Week 27 (early September) there was a steep increase in emissions that led to a peak emission rate observed in Week 33. This coincided with increasing air temperatures, an increasing pace of sludge additions, and the accumulation of sludge in the pan.

After the emission peak (Week 33) the emissions decreased over time. By Week 41 (December 2021) the emissions had dropped by approximately 62% from the peak. The decrease corresponded to an interval with no sludge additions, which likely due to the reduced availability of organic carbon in the sludge. Only 36 mm of rain fell from

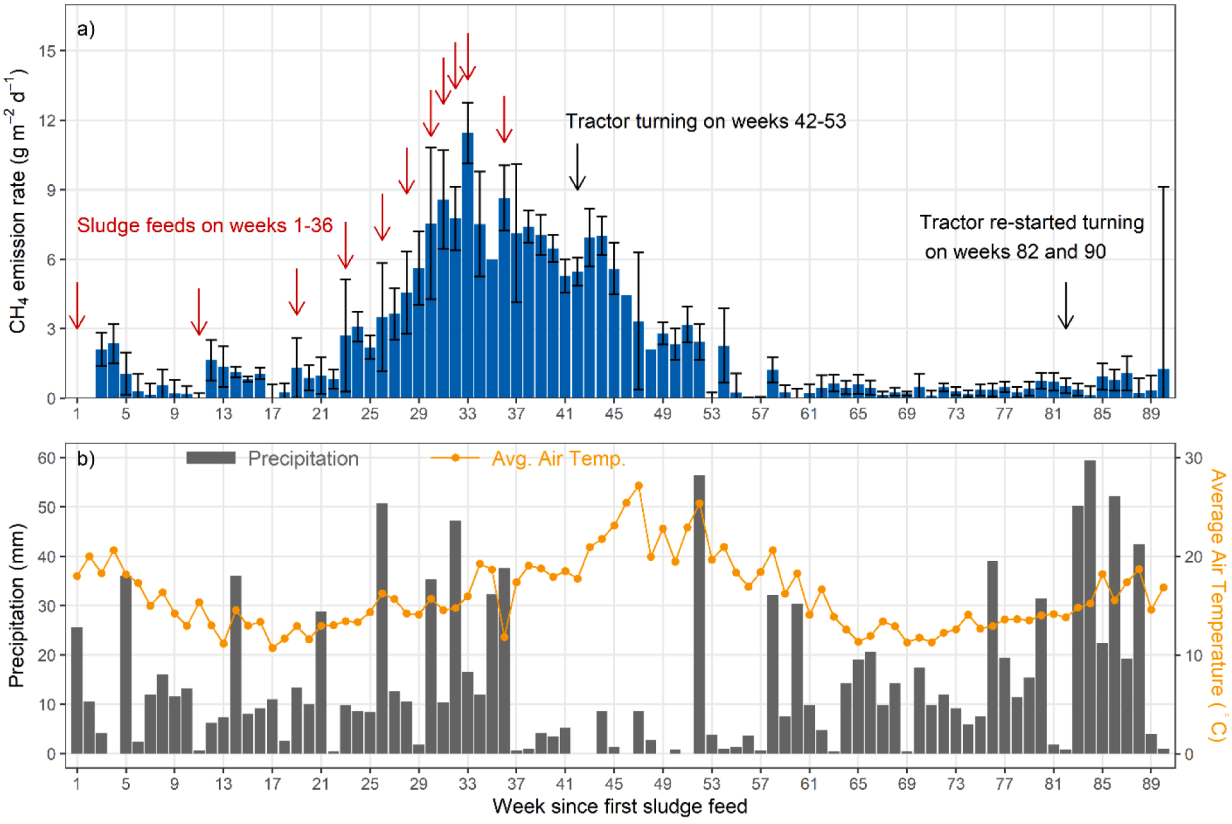

Fig. 1. Weekly CH_4 emissions from the sludge drying pan (a) and weekly total precipitation and average air temperature (b) at the SDP site over the measurement period from 24 March 2021 to 27 November 2022. The first batch of sludge was added in Week 1 and the last sludge was added in Week 36. Red arrows indicate the sludge additions. Black arrows indicate the first and the last turnings, and there were a total of 16 turning events. The error bars denote the standard deviations.

Weeks 36 to 51 (Fig. 1B), therefore rainfall events are unlikely to be a key factor contributing to reduced emissions. We note that a tractor turning sludge event on Week 41 coincided with a short-term 30% increase in the emissions over the subsequent two weeks. Emissions plateaued at a low level after Week 55. There was a slight increase in emissions when sludge turning was resumed on Week 82, with emissions rising from an average of 0.2 to 0.9 g m^{-2} d^{-1}.

We use a generalized additive model (GAM) with a normal error distribution to propagate the uncertainty of daily gas emissions over the measurement period (Bai et al. 2020, Wood 2006). Over the 90 weeks, the cumulative CH_4 emissions from the SDP was calculated to be 32.5 (± 1.07) t (± s.e.) (Fig. 2), giving rise to a mean daily CH_4 emission rate of 2.3 (± 0.08) g m^{-2} d^{-1}. The dynamic emission pattern highlights the importance of continuous (or near continuous) measurements in order to accurately quantify CH_4 emissions. This capability is an important advantage of micrometeorological measurement techniques like IDM.

CH_4 emission factor

The IPCC (2006) outline a simple procedure for estimating CH_4 emissions from wastewater treatment facilities based on chemical oxygen demand (COD) levels of the waste (Equation 1):

$$CH_4 Emissions = B_0\ MCF\ (COD_{in} - COD_{out}) \quad (1)$$

where B_0 (0.25 kg CH_4 kg^{-1} COD) is the theoretical COD to CH_4 conversion factor, MCF is a CH_4 correction factor that depends on the type of treatment, COD_{in} (kg) is the cumulative amount of COD fed to the SDP via sludge feeding, and COD_{out} is the amount of COD remaining after drying.

The accumulated COD_{in} from the sludge additions to the SDP was 1,518 t. The COD_{out} measured at Week 63 was 834 t. This duration is representative of the median pan cycle duration at site (i.e., the time at which the pan contents would typically be harvested and stockpiled). The COD_{out} measured at Week 90 (95th percentile pan cycle duration) was 472 t.

Using the COD measurements and the cumulative CH_4 emissions over 63 weeks (median operational cycle duration), the MCF for this SDP was calculated to be 0.17 (Table 1). Over 90 weeks (95th percentile operational cycle duration of all drying pans), the MCF dropped to 0.12 (Table 1). These values were slightly below the IPCC default value of 0.2 for a shallow anaerobic lagoon (defined as < 2 m in depth), but well within the expected range of 0–0.3 given by the IPCC. The MCF measured at the median cycle duration time was very close to the IPCC default value, giving confidence in the current reporting methodology, given sludge loading variability, seasonal factors, and variations in duration drying cycle from the pans.

Table 1
The chemical oxygen demand (COD) information, cumulative CH_4 emissions over 63 weeks (March 2021−May 2022) and 90 weeks (March 2021−November 2022), and the calculated Methane Correction Factor (MCF). The IPCC MCF default value is also shown.

Comparison of Methane Correction Factor (MCF)	63 weeks	90 weeks
SDP 49 annual CH_4 emissions measured (t)	30.4 ± 0.91[†]	32.5 ± 1.07[‡]
COD_{in} (t)	1518	1518
COD_{out} (t)	834	472
COD in sludge (t)	697	335
COD in supernatant decant (t)	137	137
Emissions = B_0 MCF (COD_{in} - COD_{out})		
B_0	0.25	
MCF (IPCC)	0.2	
MCF (from study)	0.17	0.12

[†] The cumulative CH_4 emission is calculated from Weeks 1 to 63.
[‡] The cumulative CH_4 emission is calculated from Weeks 1 to 90.
B_0, the theoretical COD to CH_4 conversion factor.

Overall COD balance

The COD mass balance analysis (Fig. 3A) shows that after 90 weeks, 9% of the feed COD was removed via the supernatant decanted, 22% ended up in the dried sludge, while the rest (~69%) was emitted. The "emitted" COD can be 1) converted to CO_2 via aerobic and anoxic reactions, or 2) converted to CH_4 and CO_2 via anaerobic transformations. The anaerobic processes consumed 8% of the total COD fed to the pan, estimated from the measured total CH_4 emissions. Based on the COD mass balance, the remaining "emitted" COD (61% of COD fed to the pan) should be attributed to aerobic and anoxic reactions, e.g., with oxygen/nitrite/nitrate/sulfate as the electron acceptors, enabled by the surface transfer of oxygen from the atmosphere.

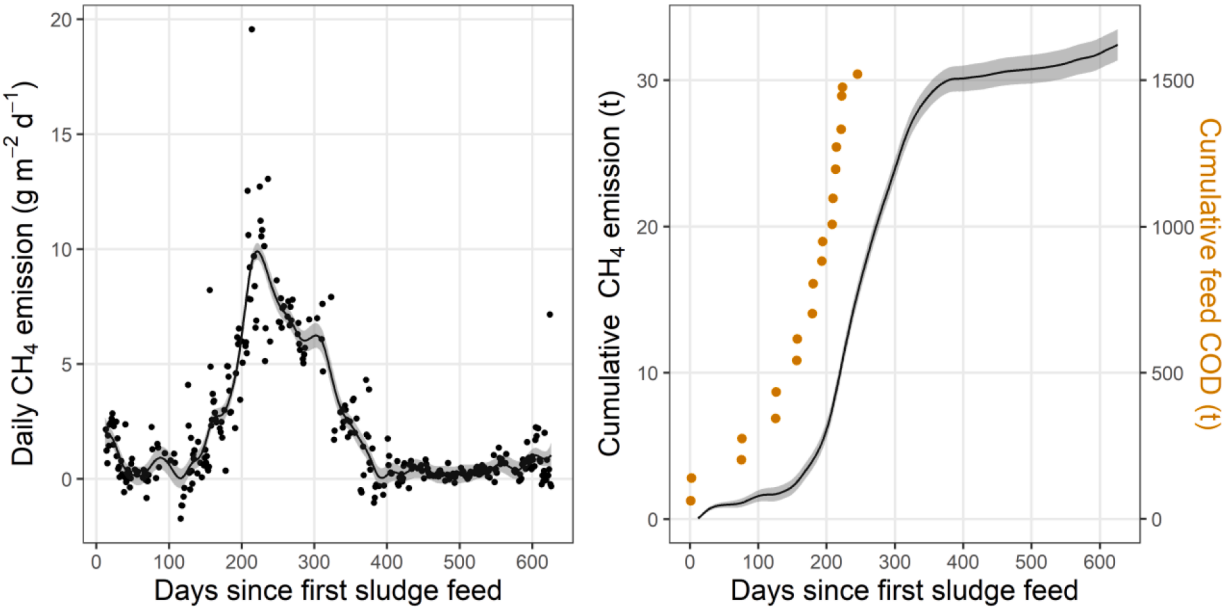

Fig. 2. Daily and cumulative CH_4 emissions from the sludge drying pan estimated using GAM (left) and cumulative COD fed to the system (right). Note, the measured daily emissions are shown in black dots and the modelled values are shown in line (left panel), the cumulative emissions are shown in line, the feeds are shown in orange dots (right panel).

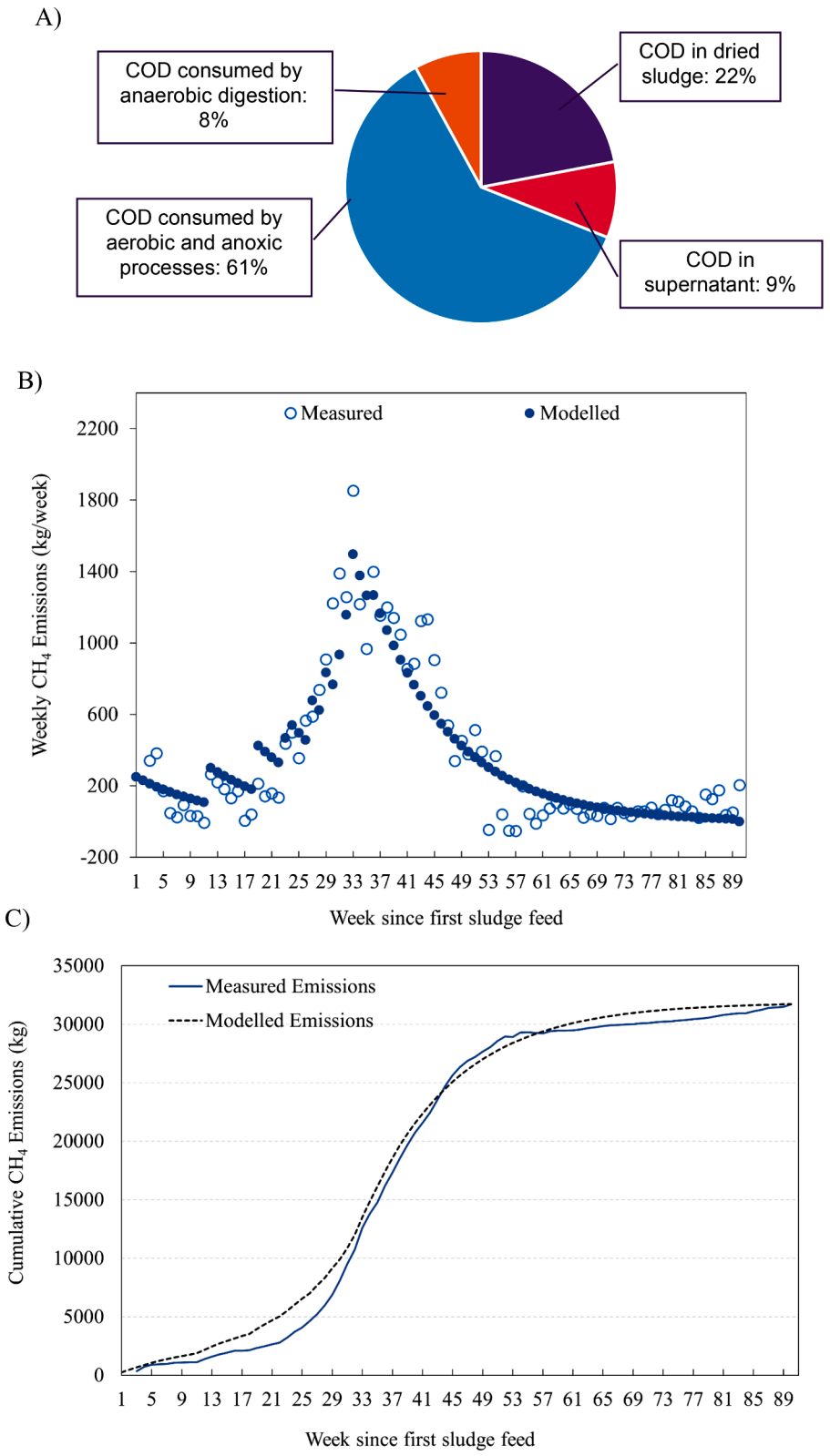

Fig. 3. Overall the COD balance of the sludge drying pan (A), measured and modelled weekly CH_4 emissions (B), and accumulative (C) CH_4 emissions from the sludge drying pan.

The observed MCF was higher at 90 weeks than it was at 63 weeks. This indicates that as the COD in the sludge reduced over time, a decreasing proportion of this reduction was from anaerobic methanogenic processes. This is likely driven by the relatively fixed rate of oxygen transferring into the sludge bed and the reducing rate of COD reduction over time as the substrate is consumed.

Modelling-based analysis and implications for mitigation strategies

In order to understand the impact of the various operating

parameters on the measured emissions and to determine the key parameters that can be used to estimate SDP emissions in the future, two model approaches were examined. The first being a first-order kinetic model relating emissions to volatile solids destruction and the second being a sediment model relating MCF to operating conditions.

Dynamic emissions fitted by a first-order kinetic model

The dynamic CH_4 emissions were modelled using the first-order kinetic equation (Equation 2), assuming volatile solid (VS) hydrolysis is the rate-limiting step.

$$\mathrm{ER}\ (t) = \mathrm{Y}\cdot\mathrm{VSD}(t) = \mathrm{Y}\cdot\mathrm{VSD}_m\cdot\left(1-e^{-kt}\right) \quad (2)$$

where ER(t) denotes the CH_4 emission on Day *t*, *Y* is the apparent biochemical CH_4 potential (calculated to be 98 kg CH_4/t volatile solid based on the measured values, i.e., the total CH_4 emissions and the overall degraded volatile solid amount over the entire operational cycle), $VSD(t)$ is the volatile solid destruction amount on Day *t*, VSD_m is the maximum volatile solid destruction (measured to be 43% of the feed VS), *k* is the hydrolysis rate coefficient (d^{-1}).

The hydrolysis rate coefficient (*k*) was estimated to be 0.013 d^{-1} by minimizing the modelled and measured CH_4 emissions. The modelled cumulative CH_4 emissions show an excellent fit with the measured data ($R^2 > 97\%$, Figs. 3B & C). The congruency indicates that the first-order kinetic model is adequate to describe the temporal variations of CH_4 emissions from SDPs, and that the rate of CH_4 emissions can be largely explained by the COD loading and timing. Given the model fit holds over an extended duration of varying weather conditions, seasonal conditions such as rainfall and air temperature were not major causes of emission variation.

Dependence of MCF values on SDP operational conditions

The MCF value for this study was estimated to be 0.17 at the median drying cycle time for the open air drying pans (Table 1). This value is comparable to, albeit slightly lower than, the value of 0.2 recommended by IPCC (2006). To further evaluate how varying SDP operating conditions (e.g., surface organic loading and temperature) influence CH_4 emissions (represented by MCF values), a one-dimensional sediment model was established. Details of the model are presented in *Supplementary Materials*. Briefly, it was found that MCF values are positively related to surface organic loadings and temperatures (Fig. S4). At a typical temperature of 20 °C, the MCF increases from 0.02 to 0.6 with the surface organic loading being elevated from 50 to 300 g COD m^{-2} (the typical operational range of SDPs, (Tchobanoglus et al. 2003)). The surface organic loading of this study is ~70 g COD m^{-2}, locating it at the low end of the operational range, which is mainly responsible for the relatively low MCF observed.

It is to be noted that the modelling results can be considered indicatively only, as the sediment model is largely simplified due to the lack of process data, such as temporal and spatial profiles of N compounds and oxygen. The sediment model has not been validated with an independent data set because of the lack of full-scale SDP monitoring data. Although further validation of the model is needed, the model provides a simple and useful tool to estimate the CH_4 emissions from SDPs operated under different temperature and surface loading conditions. A future study could be used to expand the simplified model to a more comprehensive one when relevant data are available.

Significance of this study

With the rising awareness of GHG emissions from WWTPs, GHG monitoring and quantification has been widely performed in full-scale plants (Daelman et al. 2012, Gruber et al. 2021, Kosonen et al. 2016). However, most studies focused on N_2O and CH_4 emissions from aeration tanks using online hoods to capture the emitted gases (Duan et al. 2020). However, the gas hood method is not applicable to SDP systems which are expected to have large spatial and temporal variability (Majumder et al. 2014, Pan et al. 2016). This study, to the best of our knowledge, is the first full-scale GHG monitoring campaign for an SDP lasting for the entire operational cycle. The monitoring results indeed show significant temporal variations of CH_4 emission (Fig. 1), which further highlights the importance of conducing long-term continuous monitoring for quantifying GHG emissions from SDPs.

This study demonstrates that the reported values for open air SDPs calculated using IPCC guidelines are consistent with actual measured emissions and confirms that open air SDPs are a significant source of GHG emissions. At the WWTP facility studied here (Melbourne Water), open air SDPs and biosolids stockpiles are estimated to contribute approximately 50% of the total reportable fugitive GHG emissions from the facility, responsible for 154,000 t of CO_2 equivalent (CO_2-e) against a total reported Scope 1 emissions of 290,000 t CO_2-e in 2021/22 (https://www.melbournewater.com.au/water-data-and-education/environmental-issues/our-path-net-zero. Accessed on 15 February 2021). In China, it was also reported that emission from sludge treatment and disposal accounted for over 60% of total emissions (Zhao et al. 2021). This enables wastewater facilities to make informed decisions regarding interventions to avoid these emissions, which may include: (i) enhancing the drying process to promote aerobic conditions and faster drying, and (ii) consideration of low emission thermal treatment and carbonization technologies. The results of this study provide critical information for wastewater treatment plant owners and operators that open air SDPs are a significant source of GHG emissions. As a result, alternative sludge drying technologies need to be considered as the industry works to achieve net zero. In addition to being a large source of GHG emissions, open air SDPs can also be a significant source of odour, present land and groundwater contamination risks, have a large footprint, and exhibit poor performance during wet weather.

Conclusions

Methane emission from a sludge drying pan was monitored for 90 weeks covering an entire drying cycle using an IDM technique coupled with open path CH_4 laser meausrements. A dynamic emission pattern was observed, which increased over a sequence of sludge additions, followed by a rapid decrease as sludge additions stopped. The effect of sludge turning events leading to short-term increased emissions was also observed. The dynamic pattern highlights the importance of high-frequency measurements to accurately characterize the emissions.

A CH_4 correction factor, i.e. the proportion of the organic carbon lost during the drying process that was converted to CH_4, as measured on the basis of COD, was determined to be 0.17 at 63 weeks, the median operational cycle duration. This is slightly below, but broadly consistent with, the default value of 0.2 proposed by the IPCC for shallow anerobic wastewater lagoons. Modelling-based analysis suggests the relatively low MCF is mainly attributed to the low surface organic loadings (~70 g COD m^{-2}) and the low ambient temperature (yearly average of 18 °C).

Materials and Methods

Experimental site

The experimental site was located at Melbourne Water's Eastern Treatment Plant, which is a sewage treatment plant (~1,100 ha in size), 31 km southeast of Melbourne, Victoria, Australia. Incoming domestic and industrial wastewater is treated via conventional primary and secondary treatment from which sludge is thickened and pumped to mesophilic anaerobic digesters. Digested sludge is pumped into sludge drying pans (SDP's) and dried/stabilized to produce biosolids (Fig. S1). The treatment of sludge in the SDP includes a decanting phase and evaporation phase. The median treatment period is 400 days (57 weeks),

depending on the weather. For a typical pan, the filling and decanting cycle during the decanting phase repeats 5-6 times. Once the pan has been filled to the target solids loading (~400 t ha^{-1}), the pan is left to "stand" while the supernatant is decanted for about 180 days. The standing period ends when the solids are apparent through the supernatant liquid, and clear supernatant can no longer be extracted. The pan outlets are then closed, and the sludge is left to dry until the material can be transformed to windrows (turning period, ~220 days). Windrows are turned regularly until the sludge is harvested and stockpiled onsite.

There are a total of 106 ha of open air sludge drying pans located at the treatment plant and a further 11 identical drying pans (27.5 ha) are located at the Eastern Treatment Plant's southern drying pan area. SDP 49, located at the southwest corner of this area was chosen for emission measurements. This choice facilitated emission measurements during west and southwest winds when no other SDPs were upwind of SDP 49 (the IDM measurement technique requires "isolation" from interfering gas plumes). The SDP 49 is 100 × 240 m in size, and when full, the sludge is approximately 1 m deep. The pan base is made of hardened cement treated crushed concrete and laid out in a north-south direction. The terrain surrounding the experimental site is flat and covered with short grass. The average minimum/maximum air temperature was 10.2/19.8 °C, with annual precipitation of 705 mm. The prevailing winds was SW and NE for winter and summer time, respectively (BOM 2022). A widely spaced row of small trees was located 70 m west of the pan. There were no other significant CH_4 sources west of the experimental pan (i.e. within 500 m). These characteristics mean that the site is aerodynamically simple and well suited to micrometeorological emission measurements.

The first sludge feed was added to SDP 49 on 12 and 13 March 2021. Here time is denoted as weeks since this initial sludge feed. The first two sludge fill event emissions (12–13 March 2021) were missed because the instrument was not ready. Emission measurements began on Week 3 (on 24 March 2021) and concluded on Week 90 (on 27 November 2022). Following the first sludge feed, more sludge materials were added several times between May and November 2021. Sludge interim samples (across the entire SDP 49) were taken in May and December 2022 and analyzed at a NATA accredited laboratory to determine the carbon oxygen demand value (COD), total solids and volatile solids fractions. These measurements were used to conduct a mass balance across the process and calculate a site-specific MCF factor for SDP 49. The COD and other characteristics of the added sludge are shown in Table S1. Turning of the sludge (to facilitate drying) started on Week 42 and continued once or twice per week depending on the weather (Table S2).

Measurement techniques

The IDM technique infers the emission rate of CH_4 from the SDP (Q) from the measured increase in CH_4 concentration downwind of the pond (above the upwind background, C-C_b), based on dispersion model calculations (Equation 3):

$$Q = (C - C_b)/(C/Q)_{sim} \quad (3)$$

where $(C/Q)_{sim}$ is the model calculated link between Q and (C-C_b), calculated using measured wind conditions. The WindTrax dispersion model (thunderbeachscientific.com, version 2.0.9.7) is used for the calculations.

Two open-path lasers (Gasfinder 2.0, Boreal Laser Inc., Edmonton, Canada) were used to measure CH_4 concentrations. Briefly, the sensor sends a collimated beam from a tunable infrared laser diode to a distant retro reflector mirror, from which the beam is reflected to the receiver optics and a detector. The outgoing beam is altered by CH_4 molecules over the path length (230 m, one-way path in this study), giving a measure of concentration. Concentration is measured every few seconds and averaged into 15-min intervals for analysis. The precision of CH_4 concentration is 10 ppbv (ppbv, 1 part per billion volume) at a 100 m path length (between the laser and retro reflector) (Bai et al. 2022). Figure S2 shows the experimental setup. During westerly winds (the measurement target) the laser on the west side of the SDP gives C_b, and the laser on the east side gives the downwind C.

A 3-dimensional sonic anemometer (CSAT3, Campbell Scientific, Logan, Utah, US) and data logger (CR23X, Campbell Scientific, Logan, Utah, US) provided the wind information needed for IDM. The anemometer was located at a height of 2.6 m above the ground, and the wind information was collected at a frequency of 10 Hz. A SAS software (SAS 9.4, SAS Institute Inc., Cary, NC, USA) was used to merge sonic data and OP laser data, and calculated friction velocity (u_*, m s^{-1}), atmospheric stability (L, m), and surface roughness length (z_0, m) at 15-minute averages.

Not all periods provide good emission measurements. Poor-quality periods were eliminated when either the laser measurements did not meet the manufacturers quality control thresholds (light levels < 5,000 or > 10,000; $R^2 < 0.96$), or the wind conditions did not meet common IDM criteria ($u_* < 0.1$ m s^{-1}, $|L| < 2$ m, $z_0 > 0.05$ m, wind direction was < 200° or > 315°, SDP 49 measurement area < 20%, measurement area from other pans > 5%).

CRediT authorship contribution statement

Mei Bai: Conceptualization, Methodology, Formal analysis, Writing – original draft, Writing – review & editing. **Zhiyao Wang:** Formal analysis, Writing – review & editing. **James Lloyd:** Conceptualization, Funding acquisition, Project administration, Writing – review & editing. **Dilini Seneviratne:** Conceptualization, Funding acquisition, Project administration, Writing – review & editing. **Thomas Flesch:** Conceptualization, Methodology, Formal analysis, Writing – original draft, Writing – review & editing. **Zhiguo Yuan:** Formal analysis, Writing – review & editing. **Deli Chen:** Conceptualization, Supervision, Formal analysis, Writing – original draft.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.wroa.2023.100184.

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