## Dissolved methane in anaerobic effluent: Emission or recovery?

Xiaoyuan Zhang<sup>1</sup>, Jun Gu<sup>2</sup>, Shujuan Meng<sup>3</sup>, Yu Liu (⋈)<sup>1,4</sup>

1 Advanced Environmental Biotechnology Centre, Nanyang Environment & Water Research Institute, Nanyang Technological University, Singapore 637141, Singapore

2 Department of Environmental Science and Engineering, School of Energy and Environment, Southeast University, Nanjing 210096, China

3 School of Space and Environment, Beihang University, Beijing 100191, China

4 School of Civil and Environmental Engineering, Nanyang Technological University, Singapore 639798, Singapore

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Abstract Various anaerobic processes have been explored for the energy-efficient treatment of municipal wastewater. However, dissolved methane in anaerobic effluent appears to be a barrier towards the energy and carbon neutrality of wastewater treatment. Although several dissolved methane recovery methods have been developed, their engineering feasibility and economic viability have not yet been assessed in a holistic manner. In this perspective, we thus intend to offer additional insights into the cost-benefit of dissolved methane recovery against its emission.

**Keywords** Anaerobic treatment, Municipal wastewater, Dissolved methane, Methane recovery, Carbon emission

With the fast-evolving global climate change, the energy and carbon neutral municipal wastewater treatment is under the spotlight. Different from the conventional activated sludge process and its variants which are primarily based on the concept of biological oxidation, anaerobic processes have been actively explored for direct COD capture from municipal wastewater, while maximizing the energy recovery and minimizing waste sludge generation (Liu et al., 2019; Zhang et al., 2022). However, it should be noted that anaerobic effluent contains considerable amount of dissolved methane whose release into the environment (Liu et al., 2014) would seriously compromise the energy recovery potential and contribute to significant greenhouse gas emission. As

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such, increasing effort has been devoted to developing dissolved methane recovery methods, e.g. mechanical degassing (Gu et al., 2017), membrane contactors (Li et al., 2019; Rongwong et al., 2018) etc. It should also be aware that the assessments of various recovery methods are primarily motivated by the energy recovery and consumption, without a thorough consideration of their environmental sustainability and economic viability. In this perspective, we intend to offer additional insights into the cost-benefit of dissolved methane recovery against its emission.

Given an anaerobic effluent with dissolved methane concentration of 21 g/m<sup>3</sup> (solubility of methane at 25 °C) (Li et al., 2019; Liu et al., 2014), without proper recovery, the dissolved methane will inevitably be released into the environment. In consideration of a short lifetime of methane, its 20-year global warming potential has been recommended by the Intergovernmental Panel on Climate Change (IPCC) to be 84-87 folds of carbon dioxide. Thus, this amount of dissolved methane will cause a carbon emission of  $(21 \text{ g/m}^3) \times 84=1.76 \text{ kg CO}_2\text{e/m}^3$ wastewater treated, while compromising the overall energy recovery efficiency. In fact, such a carbon emission is equivalent to the carbon emission from generating  $(1.76 \text{ kg CO}_2\text{e/m}^3)/(0.99 \text{ kg CO}_2\text{e/kWh})=1.78 \text{ kWh/m}^3$ of electricity through coal combustion, with a factor of 0.99 kg CO<sub>2</sub>e/kWh of electricity produced from coal (U.S.-Energy-Information-Administration, 2020). It is obvious that dissolved methane in anaerobic effluent is becoming a barrier towards the energy- and carbon-neutral municipal wastewater treatment if a proper measure is not in place for its recovery.

So far, several methods have been developed for dissolved methane recovery. For example, an average dissolved methane concentration of 17.1 g/m<sup>3</sup> was observed in an anaerobic effluent at 30 °C, of which

nearly 90% could be recovered by means of a mechanical degasser at an energy cost of 0.12 kWh/m<sup>3</sup> (Gu et al., 2017). Therefore, the recoverable energy could be calculated to be  $(15.4 \text{ g/m}^3)/(16 \text{ g/mol}) \times 22.4 \text{ L/mol}$  $\times 37.8 \text{ MJ/m}^3$  (methane energy content)  $\times 35\%$  (electricity conversion efficiency)/ $(3.6 \text{ MJ/kWh}) = 0.079 \text{ kWh/m}^3$ wastewater treated. Thus, the net energy utilized for degassing was estimated to be 0.041 kWh/ m<sup>3</sup> wastewater treated, which could lead to a carbon emission by  $(0.041 \text{ kWh/ m}^3) \times (0.99 \text{ kg CO}_2\text{e/kWh}) = 40.6 \text{ g}$ CO<sub>2</sub>/m<sup>3</sup>, with coal as the fuel for electrical energy production. On the other hand, the residual dissolved methane after recovery eventually resulted in a direct carbon emission of  $(1.71 \text{ g/m}^3) \times 84 = 144 \text{ g CO}_2 \text{e/m}^3$ . As such, the overall carbon emission associated with dissolved methane after recovery could be determined to be 185 g CO<sub>2</sub>e/m<sup>3</sup> wastewater treated which was only about 13% of that in the scenario of the business-as-usual (i.e. without dissolved methane recovery:  $17.1 \text{ g/m}^3 \times 84$  $=1436 \text{ g CO}_{2}\text{e/m}^{3}$ ).

In another study by Li et al. (2019), an omniphobic membrane process was proposed for harvesting dissolved methane from anaerobic effluent with a saturated dissolved methane concentration of 16.4 g/m<sup>3</sup> at 35°C. Approximately 0.04 MJ/m<sup>3</sup> of energy was needed for achieving recovery efficiencies beyond 90%, equivalent to 0.01 kWh/m<sup>3</sup>, which was close to the theoretical value reported for membrane-based methane recovery (Crone et al., 2017; Velasco et al., 2021). In this case, the energy recovered from dissolved methane could easily offset the processing energy, i.e. a net energy gain of  $(16.4 \text{ g/m}^3) \times$ 90\%/ (16 g/mol)  $\times$  22.4 L/mol  $\times$  37.8 MJ/m<sup>3</sup>  $\times$  35\%/(3.6  $MJ/kWh)-0.01 kWh/m^3 = 0.066 kWh/m^3$ , which was equivalent to a carbon offsetting of (0.066 kWh/m<sup>3</sup>) ×  $(0.99 \text{ kg CO}_2\text{e/kWh}) = 65.3 \text{ g CO}_2\text{e/m}^3$ . However, the residual methane after 90% of recovery could contribute to  $(1.64 \text{ g/m}^3) \times 84 = 138 \text{ g CO}_2 \text{e/m}^3$ , suggesting a net methane-associated carbon emission of 72.7 CO<sub>2</sub>e/m<sup>3</sup> which was only about 5.3% of that in the case where dissolved methane recovery was not practiced (i.e.  $16.4 \text{ g/m}^3 \times 84 = 1378 \text{ g CO}_2\text{e/m}^3$ ). In addition, methane solubility is inversely related to effluent temperature, indicating that the methane recovery would be more necessary at lower temperature.

It should be realized that chemicals are generally required during membrane degassing, e.g. alkaline in the omniphobic membrane process (Li et al., 2019), and the potential increases in the capital and operation costs associated with membrane degassing should also be taken into a serious account in assessing the environmental sustainability and economic viability. In fact, the dissolved methane recovery rate of membrane contactors had been reported to be 0.05 mol methane/(m²-h) (i.e. 0.8 g methane/(m²-h)) at a recovery efficiency of 96% (Velasco et al., 2021). For a middle-sized anaerobic process treating 200,000 m³/d of municipal wastewater with a 21 g/m³ dissolved methane at 25°C, the membranes needed for dissovled methane recovery would be

 $(200,000 \text{ m}^3/\text{d}) \times (21 \text{ g/m}^3)/(0.8 \text{ g/(m}^2 \cdot \text{h})) = 218,750 \text{ m}^2$ , indicating a significant increase in the captital investment and maintenance cost. In addition, membrane wetting, fouling and concentration polarization will make the operation of membrane contactors more challenging (Crone et al., 2016). Moreover, the energy required for upgrading and compressing recovered dissolved methane should also be considered, which had been reported to be about 0.011 kWh/m³ (Crone et al, 2016). Obviously, without the consideration of these factors, the energy-based assessment as currently reported in the literature, to a great extent, is misleading.

As illustrated in Fig. 1, a multiple-dimensional assessment framework of techniques for dissolved methane recovery should be exercised. For example, compared to membrane contactors, mechanical degasser would not reach the energy-neutral recovery of dissolved methane, but it has the advantages of chemical-free, simple structure, very low capital investment and operation cost with a smaller footprint. Lastly, it should be noted that the dissolved methane recovery technologies are still at the infant stage, further research is needed to make them more technologically feasible, economically viable and environmentally sustainable.



Fig. 1 Multi-dimensional assessment of techniques for dissolved methane recovery methods.

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