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## **Full-scale quantification of CH<sub>4</sub> emissions from wastewater treatment plants and biogas facilities.**

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### **1. Introduction**

Fugitive methane emissions from biogas-producing facilities have environmental, safety and economic concerns. The global warming potential of CH<sub>4</sub> seen over a 100-years cycle is 28 times higher than CO<sub>2</sub> [1]. Therefore, the amount of CH<sub>4</sub> losses from biogas facilities could strongly decrease the environmental advantages of bioenergy production compared to fossil fuels. Moreover, the uncontrolled CH<sub>4</sub> emissions result in safety problems because they increase risks of fire ignition and explosions. Finally, CH<sub>4</sub> losses results in less economic benefits in bio-methane production.

CH<sub>4</sub> emissions from biogas facilities are often difficult to quantify due to the diffusive nature of the emissions combined with large temporal variation and a challenging physical structure of a biogas facility. Only over the last few years the scientific community has developed methodologies and strategies of CH<sub>4</sub> quantifications from biogas facilities without pointing out a standardized and recognized method yet. On-site and remote sensing approaches coupled with dispersion models have been used to quantify methane emissions from biogas plants. On-site methods usually consists of leakages and losses identification followed by emissions quantification using the flux chamber technique [2], [3]. Remote sensing approaches are commonly coupled to a backward Lagrangian Stochastic inverse dispersion model where

measurements of atmospheric conditions are performed with a 3D anemometer and gas concentrations are detected using an Open Path Tunable Diode Laser Spectrometer (OP-TDLS) [4], [5].

DTU Environment at the Technical University of Denmark has adopted and further developed the dynamic Tracer Dispersion Method (TDM) for CH<sub>4</sub> quantification from large scale sources [6] such as landfills [7] and wastewater treatment plants (WWTPs) [8]. Next application of this method is intended to be at biogas facilities, which have the same features of sludge stabilization management units at most of WWTPs.

This abstract aims to describe TDM characteristics and briefly report CH<sub>4</sub> quantification from a number of WWTPs.

### **2. Method**

Total methane emissions were quantified using a mobile tracer dispersion method that combines a controlled release of tracer gas from the biogas facility with concentration measurements downwind of the facility, by using a mobile high-resolution analytical instrument [6], [7] The tracer dispersion method in general is based on the assumption that a tracer gas released at an emission source, in this case a biogas facility, will disperse into the atmosphere in the same way as methane emitted from the facility. Since the ratio of their concentrations remains constant along their atmospheric dispersion, the CH<sub>4</sub> emission rate can be calculated using

the following expression when the tracer gas release rate is known:

$$E_{CH_4} = Q_{tr} * \frac{\int_{plume\ start}^{plume\ end} (C_{CH_4}) dx}{\int_{plume\ start}^{plume\ end} (C_{tr}) dx} \frac{MW_{CH_4}}{MW_{tr}}$$

where  $E_{CH_4}$  is the methane emission in mass per time,  $Q_{tr}$  is the tracer release in mass per time,  $C_{CH_4}$  and  $C_{tr}$  are the measured downwind concentrations in parts per billion (ppb) subtracted of their background concentrations and  $MW_{CH_4}$  and  $MW_{tr}$  are the molar weights of methane and tracer gas, respectively [6]. In this study, acetylene ( $C_2H_2$ ) was used as tracer due to its long atmospheric lifetime. Downwind plume concentrations were measured driving along transects with a cavity ring down spectrometer (CRDS), which is a fast and high sensitive gas analyzer capable to detect  $CH_4$  and  $C_2H_2$  concentrations down to ppb level every second [8].

As an example of TDM application, Figure 1 depicts downwind plumes at a WWTP in Holbæk (Denmark), which has a load of about 60.000 population equivalent. A preliminary plant screening allows the identification of  $CH_4$  emission hotspots, which is used to place the tracer gas cylinders in order to insure a proper mixing of the two gases. Usually at WWTPs the anaerobic digester tanks for sludge stabilization constitute the main  $CH_4$  emission area (blue circle in Figure 1), which is where a tracer cylinder is placed (yellow triangle in Figure 1). Transverses are performed downwind the plant and  $C_2H_2$  and  $CH_4$  matching plumes are measured with the CRDS (yellow and blue plumes in Figure 1).

Mathematical and statistical elaborations are carried out for each transect. The calculation of the correlation coefficient of determination ( $R^2$  in Figure 2), where the gas concentrations are plotted against

each other, allows an additional test for plume matching.

Outcomes of the elaborations are  $CH_4$  emission rates expressed in kg/h, which represent a snapshot of whole facility release during the measuring time period.

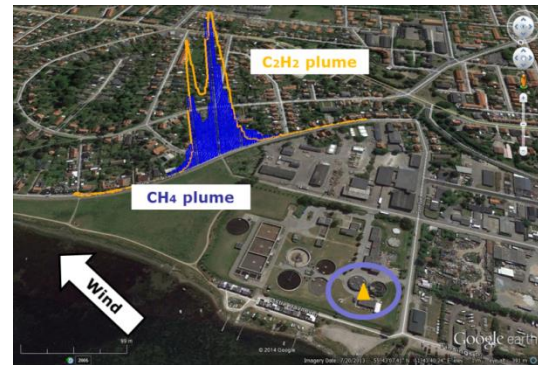


Figure 1. Methane and tracer gas plumes measured downwind Holbæk wastewater treatment plant (DK).

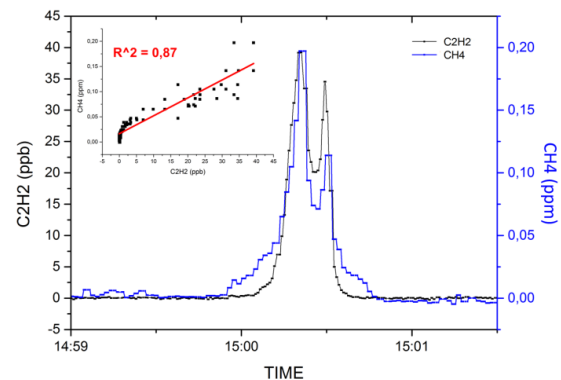


Figure 2. Data elaborations for a single plume transect measured using the Tracer Dispersion Method.

### 3. Results

In order to provide an idea about the range of  $CH_4$  emissions measurable with TDM, Table 1 reports elaborated data from different WWTPs. All of the WWTPs emitted  $CH_4$  from sludge management activities mainly the anaerobic digester tanks. At one facility, a biosolids storage area also emitted  $CH_4$ . The  $CH_4$  fluxes range from 3 to 111 kg/h underlining the

suitability of the method in quantifying both big and small emission rates.

*Table 1. Examples of CH<sub>4</sub> emission rates from WWTPs*

|         | CH <sub>4</sub> (kg/h)       | Main source                          |
|---------|------------------------------|--------------------------------------|
| Plant A | 2.9±0.5                      | Digesters                            |
| Plant B | 5.8±3.0                      | Digesters                            |
| Plant C | 18.2±6.2                     | Digesters                            |
| Plant D | West:32.4±2.6<br>East:111±46 | Sludge handling<br>Biosolids storage |
| Plant E | 37.1±16.8                    | Digesters                            |
| Plant F | 5.0 – 92.3                   | Digesters                            |

Like every CH<sub>4</sub> quantification method, also TDM has advantages and disadvantages, which are briefly listed below.

*Advantages:*

- Only one skilled operator is required;
- Straightforward data analysis and calculation when gases are fully mixed;
- Downwind plume changes can be instantaneously detected and the measurements adjusted accordingly;
- Flexibility to carry the equipment around either by car or small trolley;
- Capability to point out emissions from hot-spots;
- Possible CH<sub>4</sub> emission quantification even without locating specific hotspots;
- Identification of short time emission variation;
- Whole plant emission quantification.

*Disadvantages:*

- Dependence on favorable wind conditions combined with road access;
- Monitoring time only possible with favorable wind;
- Individual leakages are not quantified as the method integrates the whole plant emission.

The quantification of whole plant emission is considered both a pro and a con of the method because, as it on one hand can directly provide the total emission from the whole plant, on the other hand, emissions from different units cannot be quantified individually unless there is a particular plant layout where individual CH<sub>4</sub> plumes can be identified.

**5. Conclusions**

The tracer dispersion method is expected to be a suitable method for fugitive CH<sub>4</sub> quantification at biogas-producing facilities. The method has already been validated for other area sources such as landfills and wastewater treatment plants. By combining a tracer gas release with downwind plume concentration measurements using a fast and very sensitive gas analyzer, a single skilled operator can quantify CH<sub>4</sub> emission rates expressed in kg/h, which represent a snapshot of whole plant emission at the time of the measurement.

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