

Supporting information for the publication from Bühler et al. entitled “Using the inverse dispersion method to determine methane emissions from biogas plants and wastewater treatment plants with complex source configurations”

Supporting Information 1

1. Description of WWTPs

Wastewater treatment plant 1 (WWTP-1) consists of a conventional activated sludge treatment with complete nitrification and denitrification. The primary sludge passes a thickener from where it enters the digesters with a dry matter content of 4 %. The anaerobic digestion is operated at mesophilic conditions (average temperature during measurement campaign: 37.8 °C). The biogas is fed to a combined heat and power unit (CHP) for electrical power production. The heat is used for heating the digester. The excess heat is fed to a district heating network. The gas torch was never operated during the measurement campaign. After a residence time of approximately 20 days the sludge is again dewatered to 8 % of dry matter after addition of a flocculant by means of a rotary screen and then transferred to open sludge storage tanks (total volume: 1,960 m³). The sludge is regularly evacuated and transported to a larger WWTP for further treatment and disposal. Before the transport, the tanks are stirred to achieve pumpability of the sludge.

The water line at WWTP-2 (Fig2b) consists of a screen, a grit chamber, primary clarification basins and a sequencing batch reactor where the pre-treated sewage undergoes three cycles of 8 h each: (1) filling of one of the three reactors, (2) aeration, (3) sedimentation of the secondary sludge and extraction of excess sludge and discharge of the treated water into the retention basin from where it is regularly discharged into the receiving water.

The primary sludge passes a pre-thickener and is then directed to a belt thickener and enters the digesters with a dry matter content of 2 % to 3 %. The anaerobic digestion is operated at mesophilic conditions (average temperature in 2020: 37.6 °C for digester 1 and 34.4 °C for digester 2). After a residence time of approximately 20 days the sludge is again dewatered to 5 % of dry matter after addition of a flocculant by means of a rotary screen and then transferred to one of the two open sludge storage tanks (volume: 400 m³). The biogas is fed to a CHP for electrical power production. The heat is used for regulating the temperature in the digester. The sludge is regularly evacuated and transported to a larger WWTP for further treatment and disposal. The tank for sludge storage is regularly stirred in the morning. The supernatants are removed from the surface in the afternoon and filled into the storage tank for sludge water. The gas torch is rarely used, i.e. mainly to evacuate condensation water in the gas pipe system.

Table S1: Characteristics of the wastewater inflow at WWTP-1 and WWTP-2: annual values of 2019 and/or 2020 and recorded during measurement campaigns; C: concentration, COD: Chemical Oxygen Demand, NH₄-N: ammonium given as nitrogen, F: flow, MC: Measuring campaign.

| | Inflow [m ³ d ⁻¹] | C-COD [g L ⁻¹] | C-NH ₄ -N [g L ⁻¹] | F- COD [kg d ⁻¹] | F-NH ₄ -N [kg d ⁻¹] |
|----------------------------|---|-------------------------------|--|---------------------------------|---|
| WWTP-1 | | | | | |
| Average 2019 | 12268 | 293 | 33 | 3483 | 376 |
| Median 2019 | 10248 | 307 | 36 | 3377 | 368 |
| Minimum 2019 | 7829 | 156 | 11 | 1866 | 230 |
| Maximum 2019 | 30660 | 424 | 48 | 6133 | 1034 |
| Average during MC | 11168 | 299 | 36 | 2862 | 336 |
| Median during MC | 9380 | 310 | 311 | 2933 | 341 |
| WWTP-2 | | | | | |
| Average 2019 | 4458 | 240 | 34 | 1010 | 138 |
| Median 2019 | 3710 | 233 | 36 | 995 | 139 |
| Minimum 2019 | 2321 | 113 | 14 | 566 | 95 |
| Maximum 2019 | 12967 | 373 | 51 | 1757 | 168 |
| Average 2020 (01.01-30.06) | 4606 | 259 | 33 | 1126 | 137 |
| Median 2020 (01.01-30.06) | 3798 | 248 | 35 | 1009 | 139 |
| Minimum 2020 (01.01-30.06) | 2417 | 134 | 9 | 631 | 69 |
| Maximum 2020 (01.01-30.06) | 11829 | 577 | 50 | 2388 | 164 |
| Average during MC | 4005 | 270 | 37 | 1076 | 140 |
| Median during MC | 3469 | 279 | 39 | 994 | 139 |

In Table S2, the specified emission used for the source combination are given. The emissions for the sludge storage tanks were estimated based on emission rate 1.8 g CH₄ m⁻² h⁻¹ derived from data for pig slurry from Kupper et al. (2020) (i.e. baseline emissions for tank, temperate season, Supplementary data 5), a correction factor which takes into account the lower methanisation potential by 35 % for anaerobically digested slurry (VanderZaag et al., 2018) and the sludge volume from Table 1. Emission estimates for the CHP are based on emission factor of 1.74 % of the utilised CH₄ (Liebetau et al., 2013) and the gas production from Table 1, CH₄ content in biogas: 65 % (Kvist and Aryal, 2019); conversion factor of m³ CH₄ to kilograms CH₄: 0.671 (IPCC, 2006).

Table S2 Data used for source the combination of the individual sources within the WWTPs. PE = Population equivalent. The given literature data was used to define the specified emissions. Given are the emissions per area.

| Source | Literature data | Scaling | Area [m ²] | | Emission [g CH ₄ m ⁻² d ⁻¹] | |
|---------------------------------|--|---------|---------------------------|--------|--|--------|
| | | | WWTP-1 | WWTP-2 | WWTP-1 | WWTP-2 |
| Inlet | Ren et al., 2013 | PE | NA | 123 | NA | 0.3 |
| Sand trap | Czepiel et al., 1993; STOWA, 2010; Ren et al., 2013; Liu et al., 2014; Samuelsson et al., 2018 | PE | 163 | 41 | 30.8 | 39.3 |
| Primary clarifier | Ren et al., 2013 | Area | 808 | 156 | 3.4 | 3.4 |
| Activated sludge tanks | eawag, 2020 | PE | 2258 | 171 | 0.7 | 3.2 |
| Secondary clarifier | Wang et al., 2011; Ren et al., 2013; Samuelsson et al., 2018; Tumendelger et al., 2019 | PE | 1501 | NA | 0.4 | NA |
| SBR | Wang et al., 2011; Ren et al., 2013; Samuelsson et al., 2018; Tumendelger et al., 2019; eawag, 2020 | PE | NA | 1017 | NA | 0.4 |
| Thickener for primary sludge | Ren et al., 2013 | PE | 92 | 246 | 21.8 | 2.6 |
| Overflow sludge | Wang et al., 2011; Ren et al., 2013; Samuelsson et al., 2018; Tumendelger et al., 2019 | PE/Area | 320 | NA | 0.1 | NA |
| Digester | Own assumptions | other | 236 | NA | 4.2 | NA |
| Digester + CHP | own assumptions, IPCC, 2006; Liebetau et al., 2013 | other | NA | 274 | NA | 22.2 |
| Sludge storage tanks | VanderZaag et al., 2018; Kupper et al., 2020 | Area | 336 | 69 | 28.1 | 28.1 |
| Supernatants | Ren et al., 2013 | Area | 226 | 69 | 3.4 | 15.4 |
| Balloon for biogas storage | Own assumptions | other | 120 | 151 | 2.5 | 2.0 |
| CHP | IPCC, 2006; Liebetau et al., 2013 | other | 44 | NA | 216.9 | NA |

1.1 Meteorological conditions

Figure S1 shows the meteorological condition during the measurement campaign in 2019 at WWTP-1. The conditions were normal for autumn and winter although the temperatures were periodically at the upper range for the season and periods with high wind speeds occurred. The meteorological conditions during the measuring campaign in May 2020 at WWTP-2 were normal for late spring (Figure S2).

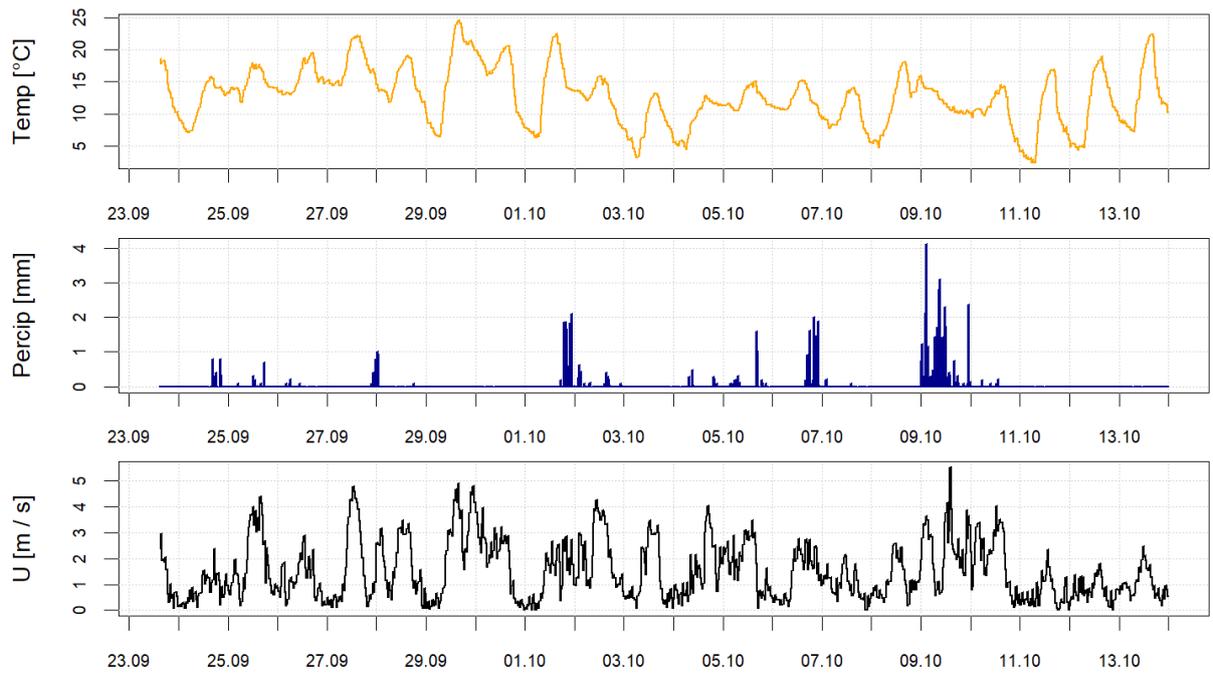


Figure S1: Overview on the temperature, precipitation, and wind speed from our own weather station during the measurements conducted at WWTP-1.

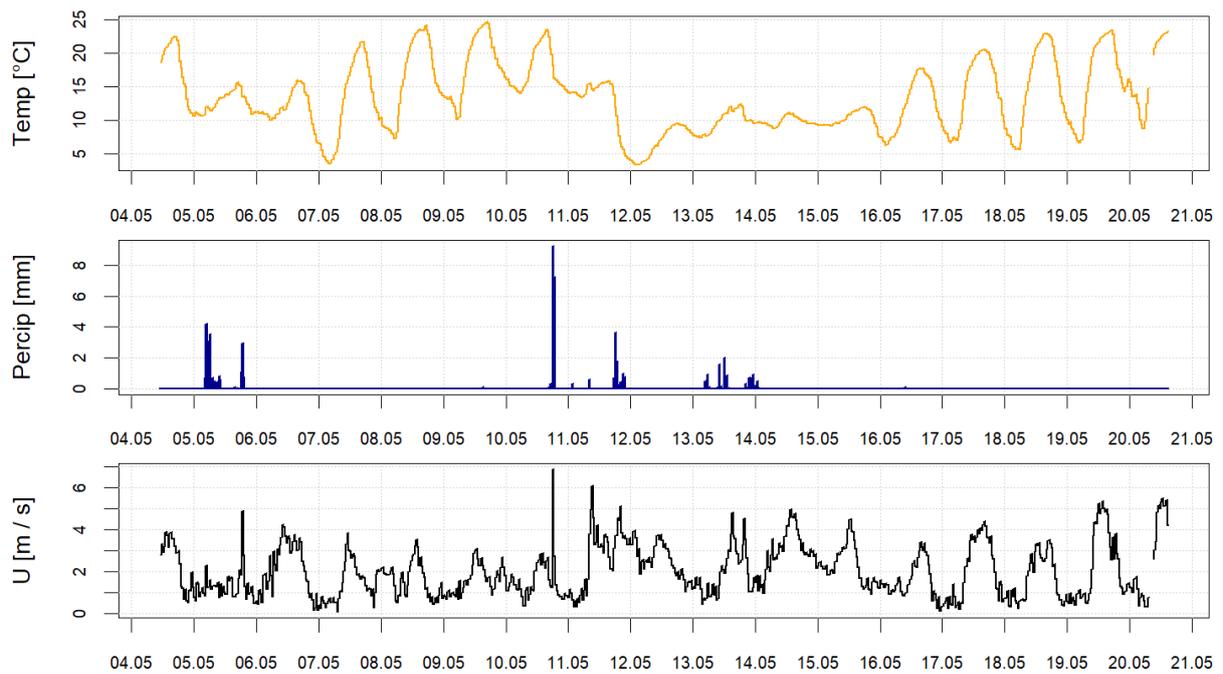


Figure S2: Overview on temperature, precipitation, and wind speed from our own weather station during the measurements conducted at WWTP-2.

2. Data filtering

Table S3 Quality filter criteria that were applied at each measurement site. It shows what kind of data was kept. N_{TD} = Number of touchdowns within the source. z_{canopy} = canopy height, A = Area of the source.

| Filter criteria | | | | | | | | | | | |
|-----------------|-------------------|--------|----------------|----------------|-------|-------|--|------------|------------|------------|------|
| Site | N of trajectories | u_* | σ_u/u_* | σ_v/u_* | C_0 | $ L $ | z_0 | N_{TD} | D/A | ΔC | WD |
| WWTP-1 | 250000 | > 0.06 | < 7 | < 7 | - | < 2 | < 0.1 | - | - | - | yes |
| WWTP-2 | 250000 | > 0.05 | < 6 | < 6 | < 10 | < 2 | < 0.1 | - | > 6.00E-05 | - | yes |
| BGP-1.1 | 50000 | > 0.05 | < 8 | < 8 | < 10 | < 2 | $z_0 > \frac{z_{canopy}}{100} \& z_0 < \frac{z_{canopy}}{3}$ | > 8.00E+04 | > 8.20E-05 | > -0.08 | yes |
| BGP-1.2 | 50000 | > 0.06 | < 8 | < 8 | < 10 | < 2 | $z_0 > \frac{z_{canopy}}{100} \& z_0 < \frac{z_{canopy}}{3}$ | > 8.00E+04 | > 8.20E-05 | > -0.08 | yes |
| BGP-2.1 | 50000 | > 0.05 | < 8 | < 8 | < 10 | < 2 | $z_0 > \frac{z_{canopy}}{100} \& z_0 < \frac{z_{canopy}}{3}$ | > 2.70E+05 | > 8.20E-05 | > -0.08 | yes |
| BGP-2.2 | 50000 | > 0.06 | < 8 | < 8 | < 10 | < 2 | $z_0 > \frac{z_{canopy}}{100} \& z_0 < \frac{z_{canopy}}{3}$ | > 2.70E+05 | > 8.20E-05 | > -0.08 | yes |
| BGP-3 | 250000 | > 0.10 | < 8 | < 8 | < 10 | < 2 | $z_0 > \frac{z_{canopy}}{100} \& z_0 < \frac{z_{canopy}}{3}$ | > 4.20E+05 | > 1.00E-04 | > -0.08 | yes |

3. GasFinder

3.1 Intercomparison of GasFinder devices

An intercomparison of the GasFinder was conducted after each WWTP campaign. This was necessary because of the offset and span between the GasFinder sensors (Häni et al., 2021). The concentrations were corrected for span and offset. As reference device the GasFinder used as background concentration (at both sites the same sensor) was used. A concentration during the campaign with wind sectors where all used GasFinders were exposed to the same background concentration was for the WWTP campaigns not possible.

For BGP-1, the concentrations were corrected with wind sectors for which all GasFinders were exposed to background concentration. For BGP-1.2, additionally a correction from an intercomparison was applied that was conducted some weeks prior to the campaign. For BGP-2.1, wind sectors were used and for BGP-2.2, an intercomparison that was conducted after the measurements. For BGP-3, the intercomparison conducted at WWTP-1 was used.

3.2 Fixation of GasFinders

Running the GasFinders out in the field can lead to a misalignment of the laser beam with the retroreflector. This often happens due to soil movement (wetting, drying, freezing, unfreezing) or wind gusts. If no automatic realignment system is available, even a daily realignment could be necessary. However, we run the devices for days without supervision and a car drive every day of several hours to the devices was not possible. Fixing the tripods with a clamp set to the ground really helped to reduce the data loss. The tripods of the retroreflectors were also fixed with a clamp set.



Figure 3 Fixation of GasFinder with clamping set during the intercomparison measurements at WWTP-2.

4. Comparison of WWTP emissions with literature data

Samuelsson et al. (2018) report an average CH₄ emission of a Swedish WWTP of 337 g PE⁻¹ y⁻¹. Delre et al. (2017) conducted measurements at five different WWTPs with average emissions between 153 g PE⁻¹ y⁻¹ and 919 g PE⁻¹ y⁻¹ and Yoshida et al. (2014) report CH₄ emission of 1339 g PE⁻¹ y⁻¹ from a WWTP in Denmark. Scheutz and Fredenslund (2019) measured emissions from several WWTPs and BGPs which were between 257 g PE⁻¹ y⁻¹ and 747 g PE⁻¹ y⁻¹ (data from four WWTPs that are not already included in Delre et al. (2017), Samuelsson et al. (2018) and Yoshida et al. (2014). Daelman et al. (2012) and Daelman et al. (2013) reported CH₄ emissions of a WWTP in the Netherlands of 306 g PE⁻¹ y⁻¹ and 390 g PE⁻¹ y⁻¹, respectively. STOWA (2010) measured emissions of three different WWTPs in the Netherlands between 140 g PE⁻¹ y⁻¹ and 310 g PE⁻¹ y⁻¹. Detailed information on the individual WWTPs is given in Table S4

The average of the 16 WWTPs reported above is 458 g PE⁻¹ y⁻¹ (median: 324 g PE⁻¹ y⁻¹). Scaled to COD in the influent, the average emissions were 0.9 % with a range of 0.3 - 1.7 %. The 16 WWTPs have a size between 40,000 and 805,000 PE and the sewage was mostly of domestic origin.

The CH₄ emissions of 166 g PE⁻¹ y⁻¹ and 381 g PE⁻¹ y⁻¹ for WWTP-1 and WWTP-2, respectively, lie within the range of the reported literature data of 140 - 1339 g PE⁻¹ y⁻¹. Compared to the literature data, the emissions of WWTP-1 are on the lower end. In terms of COD in the influent, the emissions of 0.7 % and 1.5 % lie also within the range of the reported literature. Overall, the measured emission observed in our study are in line with investigations conducted previously.

Table S4: Methane emissions per day and scaled to Population Equivalent (PE) and in percent of Chemical Oxygen Demand (COD) from the present study in from the literature

| WWTP | PE | kg CH ₄ h ⁻¹ | g CH ₄ PE ⁻¹ y ⁻¹ | % of COD | Source |
|---------------------|---------|------------------------------------|--|----------|-----------------------------|
| Moossee-Urtenenbach | 43,534 | 0.82 | 166 | 0.7 % | This study |
| Gürbetal | 14,071 | 0.61 | 381 | 1.4 % | This study |
| Göteborg | 805,000 | 31.0 | 337 | 0.6 % | Samuelsson et al. (2018) |
| Holbæk | 60,000 | 2.6 | 380 | 1.0 % | Delre et al. (2017) |
| Växjö | 95,000 | 10.0 | 919 | 1.7 % | Delre et al. (2017) |
| Källby | 120,000 | 8.6 | 628 | 1.3 % | Delre et al. (2017) |
| Lundtofte | 150,000 | 2.6 | 153 | 0.3 % | Delre et al. (2017) |
| Lynetten | 750,000 | 14.2 | 165 | 0.3 % | Delre et al. (2017) |
| Avedøre | 265,000 | 40.5 | 1,339 | NA | Yoshida et al. (2014) |
| Avedøre | 265,000 | 13.5 | 446 | NA | Scheutz, Fredenslund (2019) |
| NA | 420,000 | 12.3 | 257 | NA | Scheutz, Fredenslund (2019) |
| NA | 95,000 | 8.1 | 747 | NA | Scheutz, Fredenslund (2019) |
| NA | 125,000 | 10.0 | 701 | NA | Scheutz, Fredenslund (2019) |
| Kralingseveer | 360,000 | 12.6 | 306 | 1.3 % | Daelman et al. (2012) |
| Papendrecht | 40,000 | 1.2 | 266 | 0.9 % | STOWA (2010) |
| Kortenoord | 100,000 | 1.6 | 140 | 0.5 % | STOWA (2010) |
| Kralingseveer | 360,000 | 12.8 | 310 | 1.2 % | STOWA (2010) |
| Kralingseveer | 360,000 | 9.5 | 230 | 0.8 % | STOWA (2010) |

5. Measurement data

The processed data from all sites is given in the Supporting information 2.

6. References

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