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 m^3). 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 m^3). 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, NH_4-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 CH_4 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 CH_4 (Liebetrau et al., 2013) and the gas production from Table 1, CH_4 content in biogas: 65 % (Kvist and Aryal, 2019); conversion factor of m^3 CH_4 to kilograms CH_4: 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; Liebetrau 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; Liebetrau 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).

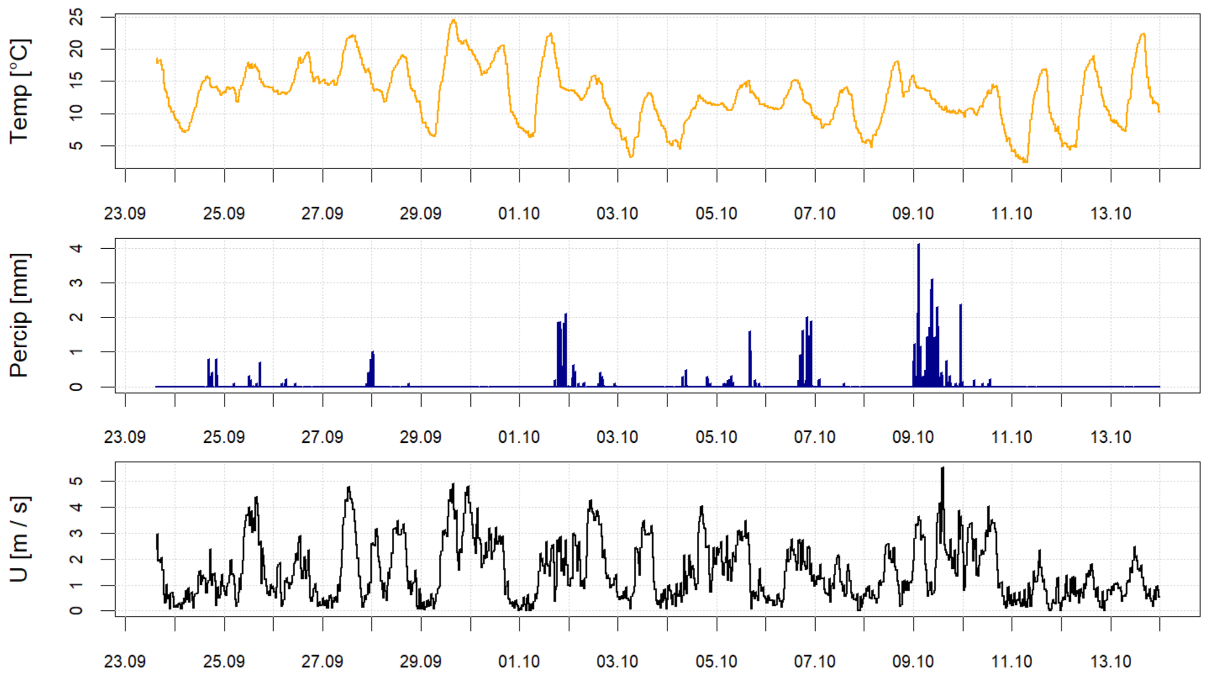

Figure S1: Overview on the temperature, precipitation, and wind speed from our own weather station during the measurements conducted at WWTP-1.

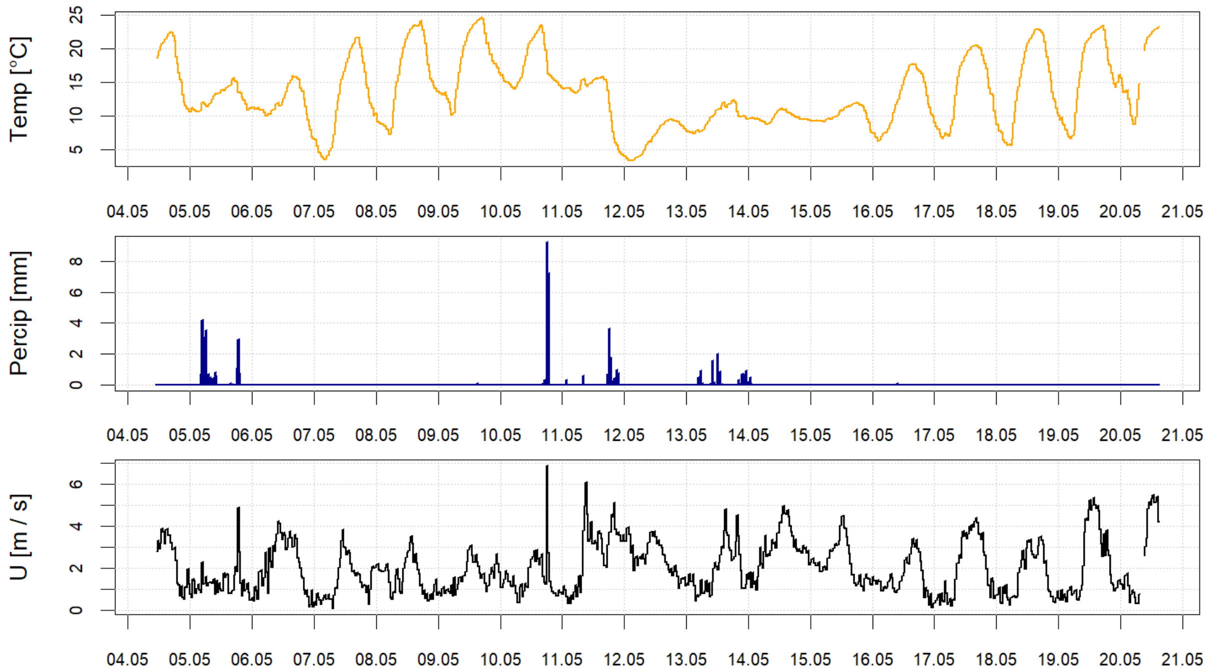

Figure S2: Overview on temperature, precipitation, and wind speed from our own weather station during the measurements conducted at WWTP-2.

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 CH_4 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 CH_4 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 CH_4 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 CH_4 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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