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Methane and ammonia emissions from wastewater treatment plants

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1. Introduction

The revised CO_2 legislation and the United Nations Framework Convention on Climate Change (UNFCCC) oblige Switzerland to regularly report the actual state of greenhouse gas (GHG) emissions (BAFU, 2018). Within the sector 5D waste management, wastewater treatment plants (WWTPs) exhibit a share of 40% of the total sector emissions and are thus a relevant source of GHGs (state in 2016[1]). The most important gas species produced in WWTPs are nitrous oxide (N_2O) and methane (CH_4). N_2O is mainly released from biological wastewater treatment and CH_4 predominantly in the sludge line (anaerobic digestion, storage) and incineration of biogas (Daelman et al., 2012; Delre et al. 2017). Within urban water management, the sewer system is also a significant source of CH_4 (Eijo-Rio et al., 2015; Mannina et al., 2018).

Emissions of ammonia (NH_3) must be reported within the Gothenburg Protocol of the UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP). Here, wastewater treatment is reported under the source 5D Wastewater handling. Data from emission measurements on CH_4 and NH_3 from WWTPs including the entire plant are not available for Switzerland. Data from other countries cannot necessarily be extrapolated to Switzerland due to possible differences in types and operation of WWTPs.

The present report presents a brief overview on wastewater treatment in Switzerland, collects actual knowledge from the literature on CH_4 and NH_3 emissions from WWTPs, sources of CH_4 and NH_3 within WWTPs, discusses the suitability of the WWTPs selected for emission measurements and provides preliminary approaches for emission estimates from WWTPs as a basis for preparing and evaluation of measurement data from field measurements. We present the results of field measurements conducted at two WWTPs in Switzerland which comprise all sources occurring within a plant. Moreover, a suggestion for measurements within WWTPs for source apportionment, namely for sludge storage tanks, based on Wagner-Riddle et al. (2006) is presented.

[1] Source: Table «Entwicklung der Emissionen von Treibhausgasen seit 1990 (April 2018)» https://www.bafu.admin.ch/bafu/de/home/themen/klima/daten-indikatoren-karten/daten/treibhausgasinventar.html (12.06.2018)

2. Wastewater treatment in Switzerland

2.1 Size and distribution of WWTPs

In Switzerland, the wastewater of 8'288'179 connected inhabitants was cleaned by 759 WWTPs in 2017. 272 WWTPs exhibited a treatment capacity of more than 10'000 population equivalents (PE). This number corresponds to 36% of the total number of WWTPs. These WWTPs treated the wastewater of 90% of the connected inhabitants[2].

2.2 The water line

Figure 1 shows a schematic picture of a typical WWTP with a conventional activated sludge system (CAS). The water line consists of a pre-treatment including physical removal of solids by a screen and a sand trap. It is followed by the primary clarifier where solids settle at the bottom of the basin. The pretreated sewage goes to the aeration tank where precipitants (e.g. FeCl, $FeClSO_4$) are added for phosphorus removal. It is then aerated to promote the growth of bacteria that consume the organics in the sewage and lead to nitrification and denitrification of the nitrogen, which occurs mainly in the form of ammonium in the sewage. In the secondary clarifier, the biomass settles and is redirected as return sludge to the head of the activated sludge tank. The residence time of the sewage in the aeration tank is in the order of several hours. A small part of the sludge removed from the secondary clarifier is redirected e.g. to the plant influent where in the primary clarifier, it is fed into the sludge treatment together with the primary sludge or goes directly to the sludge line.

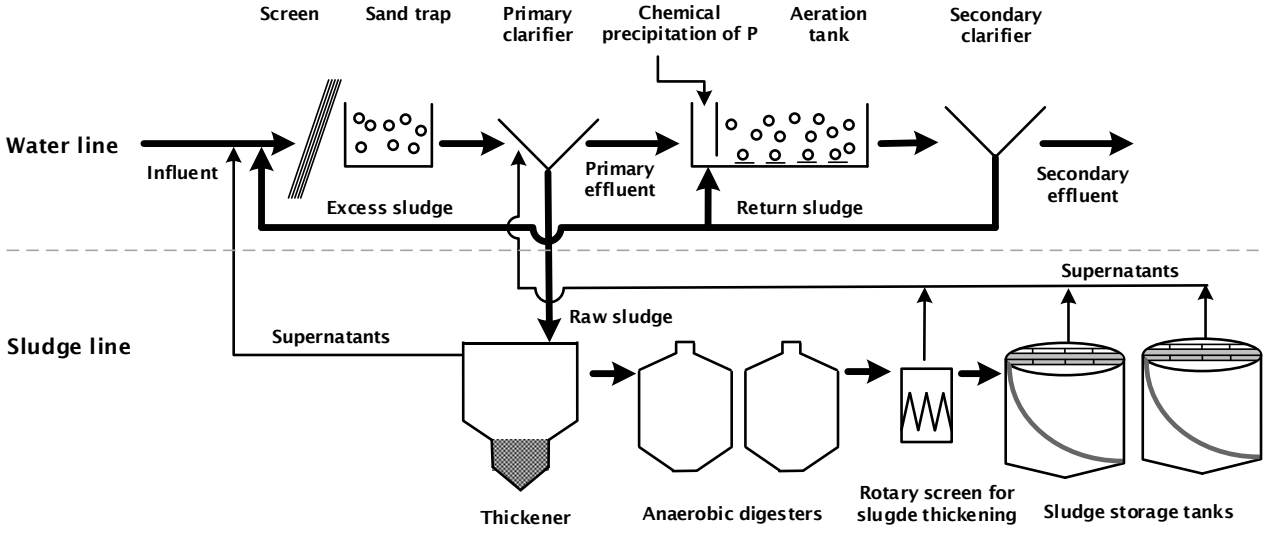


Figure 1: Schematic overview of a wastewater treatment plant.

An alternative to the CAS with continuous operation of the biological treatment is the Sequencing Batch Reactor (SBR). It is a system that becomes generally more common, also at larger WWTPs. It differs from the conventional system at the following point: the individual process steps of the biological treatment aeration and sedimentation occur in a time sequence in the same reactor. This allows to operate the duration and intensity of the single processes over a wide range and adapt them to changing conditions if needed.

Small WWTPs with <1000 PE mostly have a SBR system, i.e. about one third of Swiss WWTPs which are operated for approx. 3% of the connected inhabitants[3]. It can be assumed that the main part of the sewage is treated by either CAS or SBRs.

[2] Source: Kommunale Abwasserreinigung; URL: https://www.bafu.admin.ch/bafu/de/home/themen/wasser/fachinformationen/massnahmen-zum-schutz-der-gewaesser/abwasserreinigung/kommunale-abwasserreinigung.html (2018/11/08)

[3] Estimation based on Kupper, Chassot (1999): within the SEA monitoring net consisting of 36 WWTPs plus the catchment area. All 10 WWTPs with <2'000 PE exhibit an SBR.

2.3 The sludge line

The sludge line aims to stabilize and sanitize the sludge in order to facilitate the subsequent handling thereof and to reduce its volume for the final disposal. A side effect is the production of biogas by anaerobic digestion of the sludge, which can be used for the generation of heat and power which is sometimes denoted energy line.

The beginning of the sludge line usually consists of a thickener where a part of the water is removed from the sludge and directed to the water line of the treatment. The thickened sludge is then directed to the digester where it undergoes anaerobic degradation. The residence time of the sludge is typically 15 to 30 days (Gujer, 1999). After digestion, the sludge can be further dewatered by means of e.g a rotary screen. Dewatering is usually operated with addition of flocculants. For intermediate storage before incineration on site or transport to an incineration plant, the sludge is directed to storage tanks.

Anaerobic sludge treatment is not a priori necessary. E.g. small WWTPs with an extended aeration system (i.e. a type of SBR with a sludge residence time of approx. 20 days) obtain an aerobic stabilization of the sludge. During storage, anaerobic degradation of the volatile solids occurs due to the absence of oxygen in the bulk. However, almost all of the WWTPs >10'000 PE apply anaerobic stabilization of the sewage sludge in a digester and utilize the biogas in a combined heat and power engine (Kind, Levy, 2012). The anaerobic digestion is operated at mesophilic conditions, i.e. at temperatures around 35°C in the digester[4].

3. Literature review on methane and ammonia emissions from wastewater treatment plants

3.1 Methane emissions

3.1.1 Sources of methane emitted by WWTPs

Methane is produced in the sewer system (Fries et al., 2018; Mannina et al., 2018) and ends up in the WWTP with the sewage in dissolved form. It can be released into the atmosphere during the sewage treatment process through air stripping. Therefore, this part of methane emitted at WWTPs does not originate from wastewater treatment, but processes related to the sewer system. Production of methane occurs in the WWTP where organic carbon is present, anaerobic conditions prevail, and long sludge retention times are kept. This applies mainly for the sludge line, e.g. in sludge thickeners, during anaerobic digestion, i.e. through leakages or flaring and during storage of the anaerobically digested sludge. Methane is also emitted from the combined heat and power (CHP) engine through incomplete combustion of biogas (Liebetrau et al., 2010). CH_4 production in the water line has rarely been reported in the literature. Due to anoxic zones in the inner part of sludge flocs, Aboobakar et al. (2014) did not completely rule out generation of CH_4. Tumendelger et al. (2019) found the highest emissions of methane within the water line in the secondary clarifier and explained this by anaerobic conditions in the settled sludge.

3.1.2 Experimental approaches

To measure CH_4 emissions under real-world conditions, it can be distinguished between two different experimental approaches, such as a) flux chamber measurements and b) whole plant measurements. For a) flux chamber measurements, off-gas released at a specific part of a WWTP is captured with floating flux chambers, e.g. an aeration basin. Flux chambers are placed onto an undisturbed or a disturbed surface where air sampling takes

[4] Based on Kupper, Chassot (1999): within the SEA monitoring net consisting of 36 WWTPs plus the catchment area. All 14 WWTPs with >10'000 PE exhibit mesophilic anaerobic sludge stabilization.

place. We denote this approach "chamber measurements" (CM). For b) whole plant measurement two approaches can used: i) collection and measurement of the total off-gas of a WWTP and ii) remote sensing techniques. Total off-gas collection (i) is based on air sampling from exhaust air pipes from plants where the parts producing odors are covered, ventilated and the exhaust air undergoes a treatment. The emissions are calculated based on the concentration measured in the exhaust air and the exhaust air volume determined by e.g. anemometers. Still, not all units of WWTPs are necessarily covered and thus emissions from e.g. the secondary clarifiers are not included in the measurements (Daelman et al., 2012; STOWA, 2010). Another approach (ii) is based on remote sensing techniques such as the tracer gas dispersion method (Delre et al., 2017; Samuelsson et al., 2018; Yoshida et al., 2014). Both approaches capturing the total of emissions are denoted here "whole WWTP measurements" (WM). Published emission data differ in measurement duration. For approach (i), measurement campaigns covered durations between 8 days (STOWA, 2010) and one year (Daelman et al., 2012). Measurements based on approach (ii) imply relatively short measurement periods, i.e. mostly ≤5 measurement campaigns over ca. 1 to ≤6h (Delre et al., 2017; Samuelsson et al., 2018; Yoshida et al., 2014).

If CM are used a source apportionment is possible due to the location of sampling. However, the representativeness of the location for extrapolating the emission to the total of the treatment unit and the entire WWTP is difficult to assess and not always possible. Emission data from the sludge line are hardly available, possibly due to the difficulty to place chambers onto sludge storage tanks and piles of dewatered sludge. Whole plant methods cover the entire emission of the WWTP. A source apportionment is possible to some extent by arranging sampling in the exhaust air system (approach i) and dosing tracer gases at a specific source (approach ii).

3.1.3 Data from measurements of individual WWTP units

Eleven papers based on CM and four papers based on WM were found which provided measurement data of individual WWTP units. In most cases, data on the following process steps were available: influent pump station, sand trap, primary clarifiers, activated sludge tanks and secondary clarifiers. Not all studies investigated all WWTP units. Overall, the studies yielded 17 records. Investigated systems were CAS (13 records) and SBR and oxidation ditch (2 records each).

Table 1 shows emission data from CM and WM measurements of the water line scaled to the surface of the process unit and to the inflow at the head of the WWTP. The emissions per m^2 of a treatment unit are in the order sand trap > influent pump station > primary clarifiers > activated sludge tanks > secondary clarifiers. Scaled to cubic meter of influent, the emission pattern of the different part is as follows: activated sludge tanks > sand trap > primary clarifiers > secondary clarifiers > influent pump station. Looking at the contribution of the CH_4 emissions from individual units of WWTPs to the total emission of the waterline, the activated sludge tank and the sand trap contribute the major part. The influent pump, the primary and the secondary clarifier exhibit minor methane releases with on average less than 10% and with a maximum of 25% relative to the total emission from the water line (Table 2). The contribution of the sand trap and the activated sludge tank to the total water line CH_4 release vary largely: between less than 1% up to 67% for the sand trap and ranging from 33% to 99% for the activated sludge tank. The observed variation in emissions could be due to a different length of sewer systems occurring in the studies which could lead to a variable extent of methane formation in the sewer. Experimental artifacts could also be a reason for differing emission levels, but this point was not investigated and remains thus somewhat speculative. However, it can be concluded that non-negligible emissions of CH_4 occur in the water line with sand trap and activated sludge tank as main

sources. The influent pump, the primary and the secondary clarifier release minor amounts of methane.

Table 1: Emissions of methane from individual WWTP parts relative to the surface of a unit and to the influent. Experimental approaches are based on chamber measurements (CM) and whole WWTP measurements. (WM). The sources of the data are listed in Appendix 1, 2.1[5] for CM. WM data are based on Samuelsson et al. (2018), Daelman et al. (2012) and STOWA (2010)

*	Inf pmp	Sand tr	Pr clar	Act sl t	Se clar	Inf pmp	Sand tr	Pr clar	Act sl r	Se clar
**	CM	CM	CM	CM	CM	CM	CM,WM	CM,WM	CM,WM	CM,WM
	g CH_4 m^{-2} h^{-1}					g CH_4 m^{-3} inflow				
n	3	10	3	3	6	3	14	6	18	7
Av	1.182	2.873	0.267	1.114	0.079	0.007	0.223	0.083	0.551	0.046
Med	1.348	1.561	0.142	0.579	0.075	0.008	0.061	0.047	0.324	0.018
Min	0.496	0.202	0.020	0.064	0.005	0.001	0.000	0.024	0.003	0.000
Max	1.702	15.800	0.638	2.700	0.142	0.012	1.463	0.273	1.554	0.178

* Inf pmp: Influent pump station; Sand tr: Sand trap; Pr clar: Primary clarifiers; Act sl t: Activated sludge tanks; Se clar: Secondary clarifiers

** CM: chamber measurements; WM: whole WWTP measurements

Table 2: Proportion of methane emissions from individual WWTP units obtained by chamber methods (CM) and from Samuelsson et al. (2018). The sources of the CM data are provided in Appendix , 2.1

*	Inf pmp	Sand tr	Pr clar	Act sl t	Se clar
	Percent of total of waterline CH_4 emissions				
n	3	14	4	14	5
Av	0.3%	34%	8.3%	63%	3.2%
Med	0.4%	42%	4.1%	58%	0.1%
Min	0.0%	0.3%	0.1%	33%	0.0%
Max	0.5%	67%	25%	99%	12%

*Acronyms: Inf pmp: Influent pump station; Sand tr: Sand trap; Pr clar: Primary clarifiers; Act sl t: Activated sludge tanks; Se clar: Secondary clarifiers

Within the activated sludge system, the aerated zone produces the major part of CH_4 due to stripping of dissolved methane (Ren et al., 2013; Rodriguez-Caballero et al., 2014; STOWA, 2010; Yan et al., 2014). Emissions mostly correlate to dissolved methane in the water (Liu et al., 2014). In the aerobic zone of activated sludge tanks, the dissolved methane content is mostly low but the transfer to the air is enhanced due to aeration of the tanks. These findings were confirmed by investigations of Ren et al. (2015) in a pilot scale WWTP and by Frison et al. (2015) when investigating reject water from codigestion of sludge and digestate derived from the organic fraction of municipal solid waste at pilot scale. Daelman et al. (2012) stated that roughly 80% of the dissolved methane in the influent, of methane was oxidized in the activated sludge tank (plug flow reactor). It appeared that removal of CH_4 occurred in the aerated zone only.

Bao et al. (2016) found CH_4 emissions in biological treatment by SBR being higher by a factor of three than CAS. Nguyen et al (2019) reported in their review that CH_4 emitted from a SBR was 0.50 g m^{-3} influent and from CAS 0.18 g m^{-3} influent. Most emissions occurred in the aerobic unit or phase (Liu et al., 2014) and was caused by stripping (Bao et al., 2016). Daelman et al. (2012) observed negligible CH_4 stripped in the headworks. Most of the CH_4

[5] The complete set of records is provided in an excel sheet as supplementary information.

in the inflow was emitted in the primary clarifier and in the activated sludge treatment. Daelman et al. (2012) and Ribera-Guardia et al. (2019) observed that additional dissolved CH_4 originated from reject water produced by anaerobic sludge treatment.

Overall, there seems to be consensus that the methane emitted in the water line originates from dissolved CH_4 present in the inflow and trough addition of reject water from the anaerobic sludge treatment. CH_4 production in the secondary clarifier was claimed by one study only (Tumendelger et al., 2019).

CM measurements of the sludge line are scarce and measurements from Ren et al. (2013) are not comparable with operational conditions of Swiss WWTPs. This means that e.g. measurements from tanks for the storage of digested sludge which are common at Swiss WWTPs and assumed to be a major CH_4 source, were not found. Tauber et al. (2019) conducted measurements at an anaerobic digester at a WWTP using a chamber method and sampling from the digester's circulation pipe. They estimated emissions from the digester at 12.4 g CH_4 $PE^{-1} \cdot y^{-1}$. For further emission estimates from the sludge line, it is referred to WM data.

3.1.4 Data from whole WWTP measurements

Studies included for reporting the total of emissions of WWTPs based on WM (Table 3) are Daelman et al. (2012)[6], STOWA (2010), Delre et al. (2017), Samuelsson et al. (2018), Scheutz and Fredenslund (2019) and Yoshida et al. (2014). The 13 WWTPs which provided 16 records on CH_4 emissions from wastewater treatment are located in Denmark, Sweden and the Netherlands with a size between 40'000 and 805'000 PE. The sewage was mostly of domestic origin, but industrial wastewater occurred as well. Daelman et al. (2012) and STOWA (2010) used the approach (i) and Delre et al. (2017), Samuelsson et al. (2018), Scheutz and Fredenslund (2019) and Yoshida et al. (2014) the approach (ii) (see section 3.1.2).

Daelman et al. (2012) measured the emissions from a full-scale municipal wastewater plant over one year in the Netherlands. All the plant units were closed and ventilated. The exhaust gas was used to aerate a part of the activated sludge system. The off-gas from this unit was sampled over one year from the exhaust air pipes before an ozone washer. The individual sources were determined based on grab samples taken from the related off-gas pipes and liquid streams at five occasions over the year. The loads were determined from the measured flow rates and corresponding concentrations. Concentrations from the gas samples were determined with gas chromatography. In liquids (i.e. wastewater and sludges), the dissolved methane was measured with the salting-out method.

Delre et al. (2017) measured methane emissions at five WWTPs from Denmark and Sweden by using a tracer gas dispersion method. Gas emission rates were quantified through mobile measurement of the downwind plumes of target gases and a tracer gas. Depending on the physical size of the plant and the availability of roads in the downwind area of the plant, the measuring distance varied from 35 to 1300 m from the WWTP. Samuelsson et al. (2018) and Yoshida et al. (2014) measured at one WWTP based on the same experimental approach. Scheutz and Fredenslund (2019) reported data from WWTPs and biogas plants based on the tracer gas dispersion method. Some of the data have been published previously by Delre et al. (2017), Samuelsson et al. (2018) and Yoshida et al. (2014) and are already included in our database. To avoid redundancy, we excluded these data and only used data from four WWTPs which are not yet included in the three aforesaid studies.

[6] Additional data was published by Daelman et al. (2013): it includes the measurements at a WWTP conducted from 14 October 2010 until 26 January 2012. Daelman et al. (2012) provides the data from the same WWTP is based the data of measurements from 14 October 2010 until 28 September 2011 but the results are more detailed which allowed to calculate the data given in Table 3 which was not possible with the information provided by Daelman et al. (2013).

Average emissions were approx. 12 kg CH_4 h^{-1} (Table 3). The emissions are proportional to the plant size, i.e. the number of PE. However, the highest emission is from Yoshida et al. (2014) where the WWTP has 265'000 PE. The average emission in percent of methane in biogas produced by anaerobic digestion was 7.2% (range 2% to 15%).

Scaled to the PE of the WWTP the CH_4 emissions were on average 458 g CH_4 PE^{-1} y^{-1} (median: 324 g CH_4 PE^{-1} y^{-1}; range: 140 to 1339 CH_4 PE^{-1} y^{-1}) and to chemical oxygen demand (COD) in the influent, the emissions were 0.9% (range 0.3% to 1.7%). The two Dutch WWTPs without anaerobic sludge treatment had emissions of 266 and 140 g CH_4 PE^{-1} y^{-1}; and 0.9% and 0.5% of COD in the influent which is in the lower range. Delre et al. (2017) found the lowest CH_4 emissions at plants with enclosed sludge treatment and storage and on-site incineration of the sludge (Lundtofte and Lynetten with 153 and 165 g CH_4 PE^{-1} y^{-1} and 0.3% of COD in the influent. Enclosed storage and treatment unless the gas outflow is treated or burned does not necessarily induce low CH_4 emissions. But it was found for slurry storage that an elevated CH_4 concentration in the gas phase above the slurry surface stimulates methane oxidation (Petersen et al., 2013) which could also apply for enclosed sludge storage and contribute to a certain emission reduction. It seems likely that the on-site incineration leads to a short storage time of the sludge which results in low emissions.

The investigated Swedish plants (Källby and Växjö) exhibited the highest emissions in the study of Delre et al. (2017): 919 and 628 g CH_4 PE^{-1} y^{-1}; and 1.7% and 1.3% of COD in the influent. They have biogas upgrading systems, which are known to emit CH_4 (Reinelt et al., 2017). Furthermore, both plants stored sludge on site partly outdoors for several months. Both factors are likely the reasons for the high emissions. From Yoshida et al. (2014), even higher emissions were derived.

Delre et al. (2017), Samuelsson et al. (2018), Scheutz and Fredenslund (2019) and Yoshida et al. (2014) only provided emission numbers for individual measuring periods and not average values per WWTP. Moreover, all studies except for Daelman et al. (2012) did not conduct measurements over all seasons of the year. Nevertheless, we derived averages for the present study which is perhaps not in line with the authors intention. Therefore, the values should be used having this in mind.

Daelman et al. (2012) observed CH_4 emissions from a WWTP over the whole course of a year. The methane emission varied between 211 and 429 kg CH_4 d^{-1}. Although the emission appeared the highest during the summer months (June and July), suggesting an effect of temperature, they did not find a meaningful correlation between daily average methane emission and ambient air temperature. Possibly, the temperature of the digested sludge stored indoors was rather constant over the year and thus did not correlate with the outdoor temperature.

Table 3: Methane emissions from whole WWTP measurements (Av: average; Med: median, Min: minimum; Max: maximum). The sources of the data are provided in Appendix 1, 1.2

	Population equivalents (PE)	WWTP CH_4 total (kg h^{-1})	WWTP % CH_4 production	WWTP CH_4 (g PE^{-1} y^{-1})	WWTP CH_4 % COD in influent	WWTP CH_4 (g m^{-3} of inflow)	Proportion sludge line*
n	15	16	11	16	11	7	5
Av	267'333	11.9	7.2%	458	0.9%	4.0	48%
Med	150'000	10.0	5.9%	324	0.9%	2.4	68%
Min	40'000	1.2	2.0%	140	0.3%	1.6	2%
Max	805'000	40.5	15.0%	1339	1.7%	14	87%

* Based on: Daelman et al. (2012), Delre et al. (2017), Samuelsson et al. (2018), STOWA (2010)

Scaled to a cubic meter of inflow the emissions are on average on average 4.0 g CH_4 m^{-3} y^{-1} (median: 2.4 g CH_4 m^{-3} y^{-1}; range: 1.6 to 14.0 g CH_4 m^{-3} y^{-1}). The values at the higher and the lower end are from the same plants as mentioned above.

The average share of the CH_4 emissions between the sludge line and the water line is 48%:52%. This is likely to be biased by individual WWTPs with a high contribution of the water line (>90%). The median share is 68%:32%. The sludge line as the major source seems to be more plausible. Daelman et al. (2012) concluded that 72% of the total methane emissions came from the processes that are related to the sludge and energy line: the thickener for the primary sludge, the centrifuge, the buffer tank for the effluent of the digester (sludge residence time: 5 days), the storage tank for the dewatered sludge and methane losses from the gas engines. The buffer tank with 35% and the stock of the dewatered sludge for disposal with 15% of the total emissions were the most important individual sources. Emissions from the buffer tank were estimated at approx. 3% of the total methane produced from anaerobic digestion. The emissions related to the water line were less than 30%. STOWA (2010) found more than 90% of the total emissions coming from the water line for two WWTPs without anaerobic digestion and off transport of the sludge from the site. The high contribution of the water line to the total CH_4 emissions could be due to quasi absence of a sludge line or measurement artifacts.

Delre et al. (2017) and Yoshida et al. (2014) did not provide numbers for the water line and the sludge line. Delre et al. (2017) identified sludge treatment and energy production units as the main CH_4 emission sources. For one WWTP (Lynetten), they were able to distinguish between emissions from the water line and the sludge line at given wind conditions due to enough distance between the operational units. A proportion of 32% of CH_4 emissions produced in the water line was determined. Samuelsson et al. (2018) provide the following emissions of individual sources in percent of the total: sludge line: 81% (stockpiles of sludge dewatered by a centrifuge: 70%; exhaust air from the thickening and dewatering building: 11%); water line: 19% (sand trap: 9%, primary clarifiers: 4%, activated sludge reactors: 5%, secondary clarifiers: 2%).

An additional source is the energy line which was probably mostly included in the emissions from the sludge line because a further distinction was not possible in the studies applying the WM. Daelman et al. (2012) found a methane slip of the combined heat and power engine which occurs due to incomplete combustion of biogas of 1.3%. Woess-Gallasch et al. (2011; cited by Daelman et al., 2012) mentioned a methane slip of 1.79% as a representative value for Austrian biogas plants. Liebetrau et al. (2010) measured methane in the exhaust gas of the combined heat and power engine at seventeen German biogas plants and obtained an average methane emission of 1.73% of produced methane.

Studies which used CM and included both the water and the sludge line are from Czepiel et al. (1993), Ren et al. (2013) and Wang et al. (2011) were not included in the overview on whole WWTP emissions here because the measurements at different units of the sludge line were incomplete (e.g. did not capture leakages from the digesters or methane slip from the combined heat and power engine).

3.2 Ammonia emissions

As for methane, data for ammonia were compiled. However, data for ammonia emissions from WWTPs are even sparser. Based on Samuelsson et al. (2018), an emission of 4.3 g NH_3 PE^{-1} y^{-1} can be estimated for the whole WWTP. Sutton et al. (1995) estimated yearly emissions of a WWTP with 165'000 connected inhabitants at 27 g NH_3 per connected inhabitant. This value is based on several assumptions and was considered as uncertain by Sutton et al. (1995). Dai et al. (2015) investigated the emissions of NH_3 from municipal wastewater and

compared the outcomes with different liquid livestock manures at a laboratory scale. Emissions from wastewater were in the order of 0.003 g NH_3 m^{-2} h^{-1} and lower by a factor of 10 to 100 as compared to the manures. Samuelsson et al. (2018) estimated the contribution of the sludge line (i.e. emissions from the building where the sludge is thickened and dewatered and stockpiles of dewatered sludge) to the total emissions at 66%. As for methane, the sludge line is expected to be the main source of ammonia emissions at a WWTP.

4. Wastewater treatment plants selected for methane and ammonia emission measurements

The HAFL applies an inverse dispersion method (IDM) based on a backward Lagrangian stochastic (bLS) model combined with concentration measurements using open-devices placed up- and downwind of a stationary emission source (Flesch et al., 2005). This is a WM approach (see section 3.1.2) which we use for the determination of methane and ammonia emissions from farms, livestock housings, biogas plants and WWTPs. It has been evaluated with parallel measurements at an experimental housing (Bühler et al., 2021) where an in-house tracer ration method is applied (Mohn et al., 2018) which is considered as a reference method (Ogink et al, 2013). The applied inverse dispersion method has shown to accurately determine methane emissions at this dairy housing (Bühler et al., 2021).

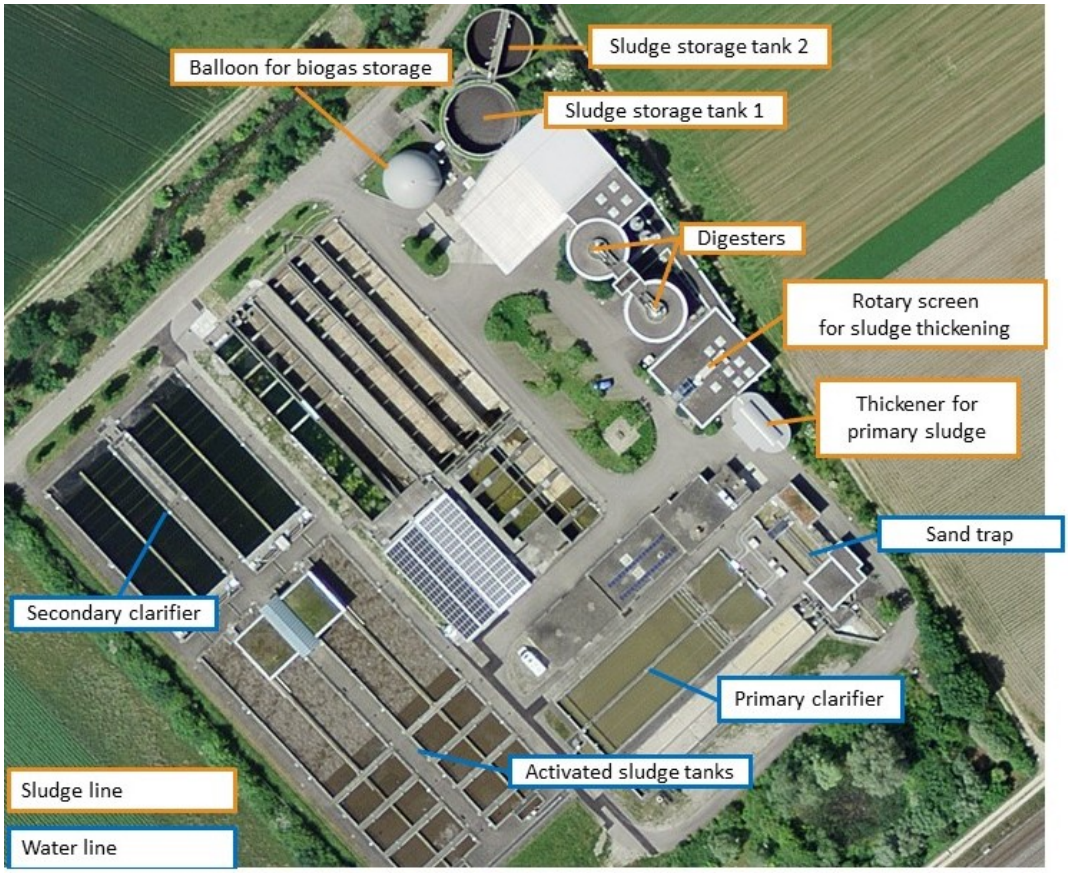
The applicability of the IDM depends on the surrounding area of the WWTP where complex terrain with slopes and obstacles like trees in the surrounding area must be avoided. The two selected WWTP Moossee-Urtenenbach and WWTP Gürbetal were selected which largely comply with these requirements (Bühler et al., 2021).

WWTP Moossee-Urtenenbach consists of a conventional activated sludge treatment with complete nitrification and denitrification. The sludge is treated through anaerobic digestion operated at mesophilic conditions. The biogas is fed to a gas motor 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 is rarely operated. The sludge is stored in open storage tanks (total volume: 1960 m^3) and regularly transported to the WWTP Bern-Neubrück for further treatment and disposal. Before the transport, the tanks are stirred in order to maintain the pumpability of the sludge.

Expected methane sources of major importance at WWTP Moossee-Urtenenbach are within the water line the aerated sand trap, the activated sludge tanks, and within the sludge line the storage tanks, the digesters, the gas storage (if leakages occur) and the combined heat and power engine, the thickener of primary sludge and the building where stabilized sludge thickening occurs.

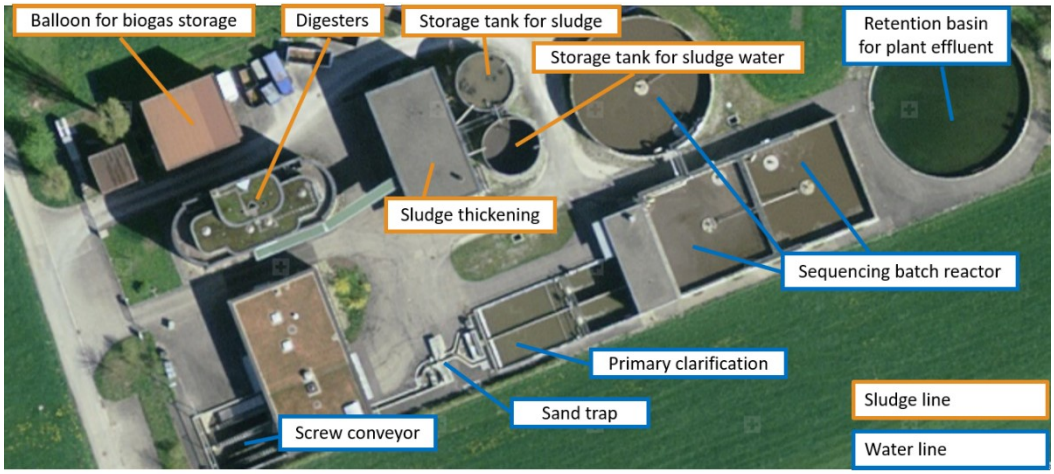
The water line at the WWTP Gürbetal consists of a screen, a grit chamber, primary clarification basins and a sequencing batch reactor where the pretreated sewage undergoes three cycles of 8 h each: (1) filling of one of the three reactors, (2) aeration, (3) sedimentation of the secondary sludge and extraction of excess sludge and discharge of the treated water into the retention basin from where it is regularly discharged into the receiving water. The sludge is treated through anaerobic digestion operated at mesophilic conditions. The sludge is stored in an open storage tank (volume: 400 m^3) and regularly transported to the WWTP Bern-Neubrück for further treatment and disposal. The tanks are stirred almost daily in the morning. The gas torch is rarely used.

Expected methane sources of major importance at WWTP Gürbetal are within the water line the aerated grit chamber, and within the sludge line the storage tanks, the digesters, the gas storage (if leakages occur) and the combined heat and power engine, and the building with stabilized sludge thickening.



Source: map.geo.ch; https://s.geo.admin.ch/7e153ef227

Figure 2: WWTP Moossee-Urtenenbach: overview on the wastewater and sludge treatment units.



Source: map.geo.ch; https://s.geo.admin.ch/7e166c8b3d

Figure 3: WWTP Gürbetal: overview on the wastewater and sludge treatment units.

The selected WWTPs are typical for Switzerland. The conventional activated sludge system is the most widely applied system for wastewater treatment (Maurer et al., 2012). SBR is also frequently used (section 2.2). Medium WWPTs with a size between 10'000 and 50'000 inhabitants as the two selected plants treat the wastewater of 42% of the Swiss population[7].

[7] Source: Kommunale Abwasserreinigung; URL: https://www.bafu.admin.ch/bafu/de/home/themen/wasser/fachinformationen/massnahmen-zum-schutz-der-gewaesser/abwasserreinigung/kommunale-abwasserreinigung.html (2018/11/08)

It has to be considered that both plants transport the final sludge to a large WWTP in the region (WWTP Bern-Neubrück) where the sludge is dewatered (usually with a centrifuge) to a dry matter content of 30% to 40% which is stored on-site for a certain time and incinerated with or without precedent thermal drying. At such plants, additional emissions are expected to occur which are partly induced by sludge from surrounding WWTPs and should, to be precise, be redistributed to these plants. Distinct measurements of methane emissions from storage tanks and/or stockpile of dewatered sludges would be necessary to estimate these emissions (Appendix 2).

5. Emission data from whole WWTP measurements in Switzerland

Methane emissions were measured at the WWTP Moossee-Urtenenbach and WWTP Gürbetal in 2019 and 2020. Ammonia emissions were additionally measured at the WWTP Moossee-Urtenenbach. The average emissions and the standard error from the measurement campaign at WWTP Moossee-Urtenenbach are 0.82 ± 0.06 kg CH_4 h^{-1} and 0.11 ± 0.06 kg NH_3 h^{-1} and for WWTP Gürbetal 0.61 ± 0.03 kg CH_4 h^{-1}. Based on Bühler et al. (2021), we estimate an uncertainty of these emission (CH_4 and NH_3) data in order of approx. 30%.

Normalized emissions for the WWTP Moossee-Urtenenbach are 166 g CH_4 PE^{-1} y^{-1} and for WWTP Gürbetal 381 g CH_4 PE^{-1} y^{-1}. Emissions scaled to COD in the influent, are 0.7% of COD and 1.4% of COD, respectively, for these WWTPs.

6. Comparison of emission factors for emission inventory reporting

Normalized emissions based on the Swiss NIR Submission of 2020 (FOEN, 2020) used for emission modeling within UNFCCC are displayed in Table 4. We also show a country specific approach proposed by the Eawag (Luck et al., 2018) which has been presented as an improved procedure for emission reporting. Both are based on data for the year 2016 and are denoted NIR subm. 2020/2016 and Country sp. prop. 2016. We compare these numbers with the normalized emissions from the measurements conducted at WWTP Moossee-Urtenenbach and WWTP Gürbetal and data from the literature.

NIR subm. 2020/2016 yields 723 g CH_4 PE^{-1} y^{-1}. Assuming that emissions from the WWTP are from 'sewage sludge & gas' released at the WWTP, the country sp. prop. 2016 results in 246 g CH_4 PE^{-1} y^{-1}, respectively. Numbers resulting from our Swiss WWTP measurements are 166 and 381 g CH_4 PE^{-1} y^{-1}. These data are within the range of the literature review which exhibit average emissions of 458 g CH_4 PE^{-1} y^{-1} (median 324 g CH_4 PE^{-1} y^{-1}) and a range between minimum and maximum of 140 to 1339 g CH_4 PE^{-1} y^{-1}. The difference between NIR subm. 2020/2016 and Country sp. prop. 2016 by a factor of about three is due to the fact that NIR subm. 2020/2016 attributes all emissions to the WWTP while Country sp. prop. 2016 includes only CH_4 from 'sewage sludge & gas' released at the WWTP. The numbers of Country sp. prop. 2016 better agree with the numbers of our Swiss WWTP measurements with a difference by 28% and 71%, respectively. The difference to the literature data is 86% (average) and 32% (median). The relatively good congruency of Country sp. prop. 2016 with numbers based on measurements is plausible since both include only emissions from the WWTP. However, it has to be noted that country sp. prop. 2016 implies additional emissions of 657 g CH_4 PE^{-1} y^{-1} from the sewer system, denoted 'CH4 from wastewater in sewer / WWTP' (Luck et al., 2018). A part thereof should be allocated to the WWTP, i.e. the water line. Luck et al. (2018) do not specify the share released from the sewer system and from the water line. In a follow-up project (MSc thesis Tobias Bührer), it could be shown that the water line does emit low emissions of methane. This is well in line with our measurements and literature. A revised country specific approach will probably result in somewhat higher emissions scaled to PE than calculated here.

Emission numbers scaled to COD are 0.6% of COD for Country sp. prop. 2016, 0.6% and 1.2% of COD for Swiss WWTP measurements and 0.9% for the literature data. We conclude that the Country sp. prop. 2016 seems to reflect data obtained from measurements fairly well while NIR subm. 2020/2016 overestimates emissions. However, NIR subm. 2020/2016 and Country sp. prop. 2016 differ less if for the latter emissions from the sewer system and from WWTPs are added up. The remark related to a revised country specific approach (see above) also applied for emissions scaled to COD.

Table 4: Methane emissions scaled to Population Equivalent (PE) and in percent Chemical Oxygen Demand (COD) derived from modeling approaches for emission reporting within UNFCCC, from whole WWTP measurements in Switzerland and the literature review

*	Modeling approaches for emission report-ing within UNFCCC		Whole WWTP measurements in Switzerland		Literature review **	Modeling appr. UNFCCC	Whole WWTP measurements in Switzerland		Literature review**
*	NIR subm. 2020/ 2016	Country sp. prop. 2016*	WWTP M.-U.	WWTP G.		Country sp. prop. 2016*	WWTP M.-U.	WWTP G.	
		WWTP CH_4 (g PE^{-1} y^{-1})				WWTP CH_4 (% COD in influent)			
Av	723	246	166	381	458	0.6%	0.7%	1.4%	0.9%
Med		-	-	-	324	-	-	-	0.9%
Min		-	-	-	140	-	-	-	0.3%
Max		-	-	-	1339	-	-	-	1.7%

*Acronyms of the Table's header row:

NIR subm. 2020/2016: *NIR Submission of 2020 for the year 2016 (FOEN, 2020) corresponding to «Corr. CH_4 application» in Luck et al. (2018) as described in chapters 3.2, 4.2, 5.2 and 6.2.*

Country-sp. prop. 2016: *country specific proposal by Eawag for the year 2016 corresponding to «Proposed CH_4 meth" in Luck et al. (2018) which differentiates emissions between sewer, WWTP and sewage gas (see chapter 8.2).*

WWTP M.-U.: WWTP Moossee-Urtenenbach

WWTP G. WWTP Gürbetal

** Data from Delre et al. (2017), Samuelsson et al. (2018), Scheutz, Fredenslund (2019), Yoshida et al. (2014) who applied a tracer gas dispersion method and Daelman et al. (2012), STOWA (2010) who applied a mass balance method. For scaling to PE: n=16 and to COD: n=11.

7. Measurements of individual sources at WWTPs

Given the fact that the mass of sludge inheres the largest amount of volatile solids and nitrogen, the sludge line is likely to be the main source of CH_4 and NH_3-emissions at WWTPs. This was already stated in section 3.1.4 and 3.2 based on data from Samuelsson et al. (2018). The energy line can produce additional CH_4 emissions due to leakages. At WWTPs such as Moossee-Urtenenbach and Gürbetal where liquid sludge is stored the storage tanks are expected to be the main source. We thus conclude that an additional measurement of emissions from sludge storage tanks would generate emission data from the prevailing source. Such measurements combined with an inverse dispersion method which provides emission from the entire plant would be useful to discriminate the emissions from sludge storage from the other sources of a WWTP. Considerations on how such measurements could be conducted are provided in the Appendix 2.

8. References

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Appendix 1
Methane emissions from wastewater treatment plants

1.1 Data from measurements of individual WWTP units

Aboobakar, A., Jones, M., Vale, P., Cartmell, E., Dotro, G. 2014. Methane Emissions from Aerated Zones in a Full-Scale Nitrifying Activated Sludge Treatment Plant. Water Air Soil Pollut. 225(1).

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1.2 Data from whole WWTP measurements

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Daelman, M.R.J., van Voorthuizen, E.M., van Dongen, U.G.J.M., Volcke, E.I.P., van Loosdrecht, M.C.M. 2012. Methane emission during municipal wastewater treatment. Water Res. 46(11): 3657-3670.

Delre, A., Monster, J., Scheutz, C. 2017. Greenhouse gas emission quantification from wastewater treatment plants, using a tracer gas dispersion method. Sci. Total Environ. 605: 258-268.

[8] This study was excluded because it is redundant to Daelman et al. (2012): it includes the measurements conducted from 14 October 2010 until 26 January 2012. Daelman et al. (2012) is based the data of measurements from 14 October 2010 until 28 September 2011 but the results are more detailed which allowed to calculate the data given in Table 3.

Samuelsson, J., Delre, A., Tumlin, S., Hadi, S., Offerle, B., Scheutz, C. 2018. Optical technologies applied alongside on-site and remote approaches for climate gas emission quantification at a wastewater treatment plant. Water Res. 131: 299-309.

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Appendix 2

Measurements within WWTPs for source apportionment based on Wagner-Riddle et al. (2006)

For emission measurements within an operation with multiple sources, the method according to Wagner-Riddle et al. (2006) could be applied. Due to the complex situation within the WWTPs, improved variants thereof are drafted.

Variants	1. Minimum	2. Good	3. Better	4. Optimum
Denomination of variants	Wagner-Riddle et al. (2006)	Wagner-Riddle et al. (2006)+	Mass balance	'Complete' mass balance
Method	Integrated Horizonal Flux (IHF)		Mass Balance Method (MBM)	
Remark regarding the method	'normal' IHF	IHF with better resolution in the downwind sector	High resolution for simplified MBM	'Complete''MBM with 3D wind measurements at C-profiles
Number of concentration profiles	4	>4	≥8	≥8
Number of concentration measurements per profile (number of heights)	4	≥4	≥4	≥4
Number of wind-profiles	1	1	1	≥4
Number of wind measurements per profile (number of heights)	4	≥4	≥4	≥4
Type of device for wind measurements	Cup/2D-Sonic	Cup/2D-Sonic	Cup/2D-Sonic	Cup/2D-Sonic/ 3D-Sonic
Total concentration measurements	16	>16	≥32	≥32
Total wind measurements	4	≥4	≥4	≥16
Minimal duration of measurement per measurement point	30sec / 5min	30sec / 5min	30sec / 5min	30sec / 5min
Minimal number of devices for concentration measurements*	2	>2	≥4	≥4

*for the type X-STREAM X2XF - Field Housing Gas Analyzer used by Eawag

The IHF method can be considered as a simplified mass balance method. Due to the expected complex emission situations at the WWTPs, we consider variant 1 (according to Wagner-Riddle et al., 2006) as feasible but rather limited and therefore suggest to apply the improved variants 2 to 4.

However, even the feasibility of variant 1 based on the available equipment (devices for concentration measurements supplied by Eawag and ev. Agroscope, cup anemometers by AWEL) seems be questionable since it competes with the aim of Eawag to measure several WWTPs in parallel. Equipment to be provided in any case: masts, intake pipes, valve manifold unit(s), vacuum pumps, air flow meter(s), control unit. The related costs for equipment are estimated at approx. CHF 20'000 (given available devices for concentration measurements can be used) and for labor at approx. CHF 50'000.

Costs for equipment or variants 2 to 4 are probably in the range of CHF 100'000 to >> CHF 100'000 (higher end for variant 4). Labor costs are expected to be in a range of CHF 100'000 to >CHF 100'000.