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Methane and ammonia emissions from wastewater treatment plants

A brief literature review

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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 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 (Delre et al. 2017). 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 are not available for Switzerland according to our knowledge. 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 on methane and ammonia emissions from wastewater treatment plants, discusses the representativeness of the WWTPs selected for emission measurements and provides preliminary suggestions for emission modeling within UNFCCC. Moreover, a suggestion for measurements within WWTPs for source apportionment 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. The water line consists of a pre-treatment including physical removal of solids by a screen and a grit removal. 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 phoshorous 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 (mainly in the form of ammonium) in the sewage. In the secondary clarifier, the biomass settles and is redirected as return sludge to the inflow 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.

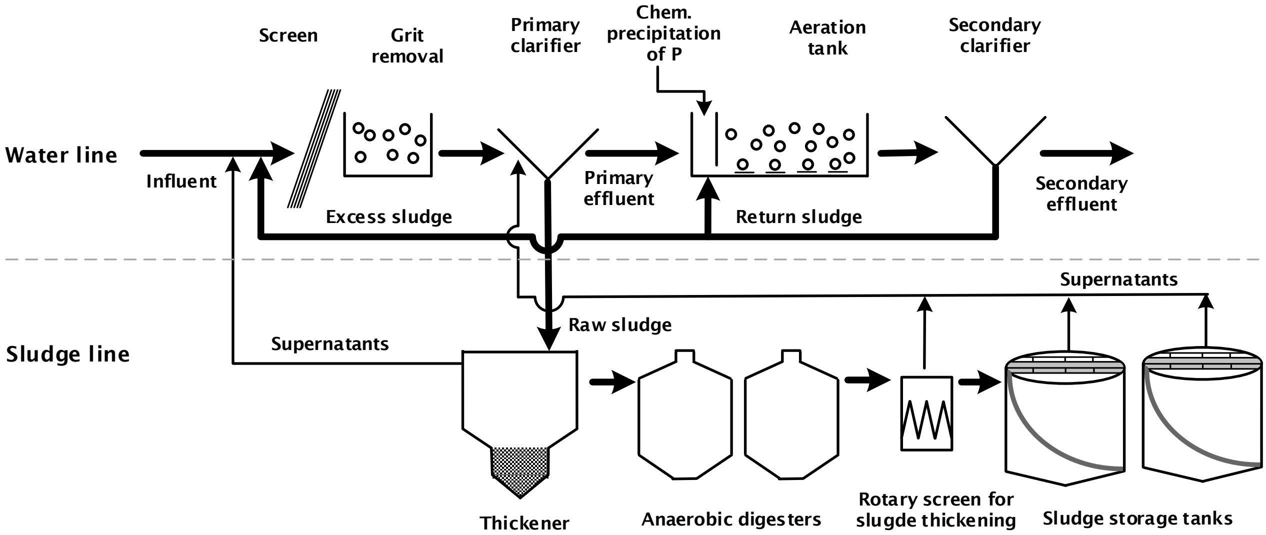

Figure 1: Schematic overview of a wastewater treatment plant.

An alternative to the conventional activated sludge system with continuous operation of the biological treatment is the Sequencing Batch Reactor (SBR). It is a system that becomes 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

[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

the main part of the sewage is treated by either conventional activated sludge systems or SBRs.

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.

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 tipically 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 final storage, 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. Wastewater treatment plants selected for methane and ammonia emission measurements

The experimental setup of the field studies comprises gas concentration measurements of CH_4 upwind and downwind of the WWTPs, measuring of wind speed and turbulence statistics of the atmosphere using 3D sonic anemometers and the recording of the geometric setup of the source area(s) as well as the measuring instruments. In combination, they constitute the input parameters for the backward Lagrangian stochastic (bLS) model yielding the emissions. Ancillary parameters obtained from the plant operators will serve as additional information source contributing to the evaluation and interpretation thereof.

The applicability of the bLS mode depends on the surrounding area of the WWTP. Complex terrain with slopes and obstacles like trees in the surrounding area must be avoided (Flesch et al., 2005). A situation with a prevailing wind direction is also advantageous. Sites with important emission sources of NH_3 and/or CH_4 in the proximity should be avoided.

The two sites WWTP Moossee-Urtenenbach and WWTP Gürbetal largely comply with these requirements (see Figure 4 and Figure 5 in the Appendix 1). An aerial view showing the parts of wastewater and sludge treatment as the expected sources of methane are provided in Figure 2 and Figure 3.

WWTP Moossee-Urtenenbach 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. The biogas is fed to a gas motor for electrical power

[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.

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.

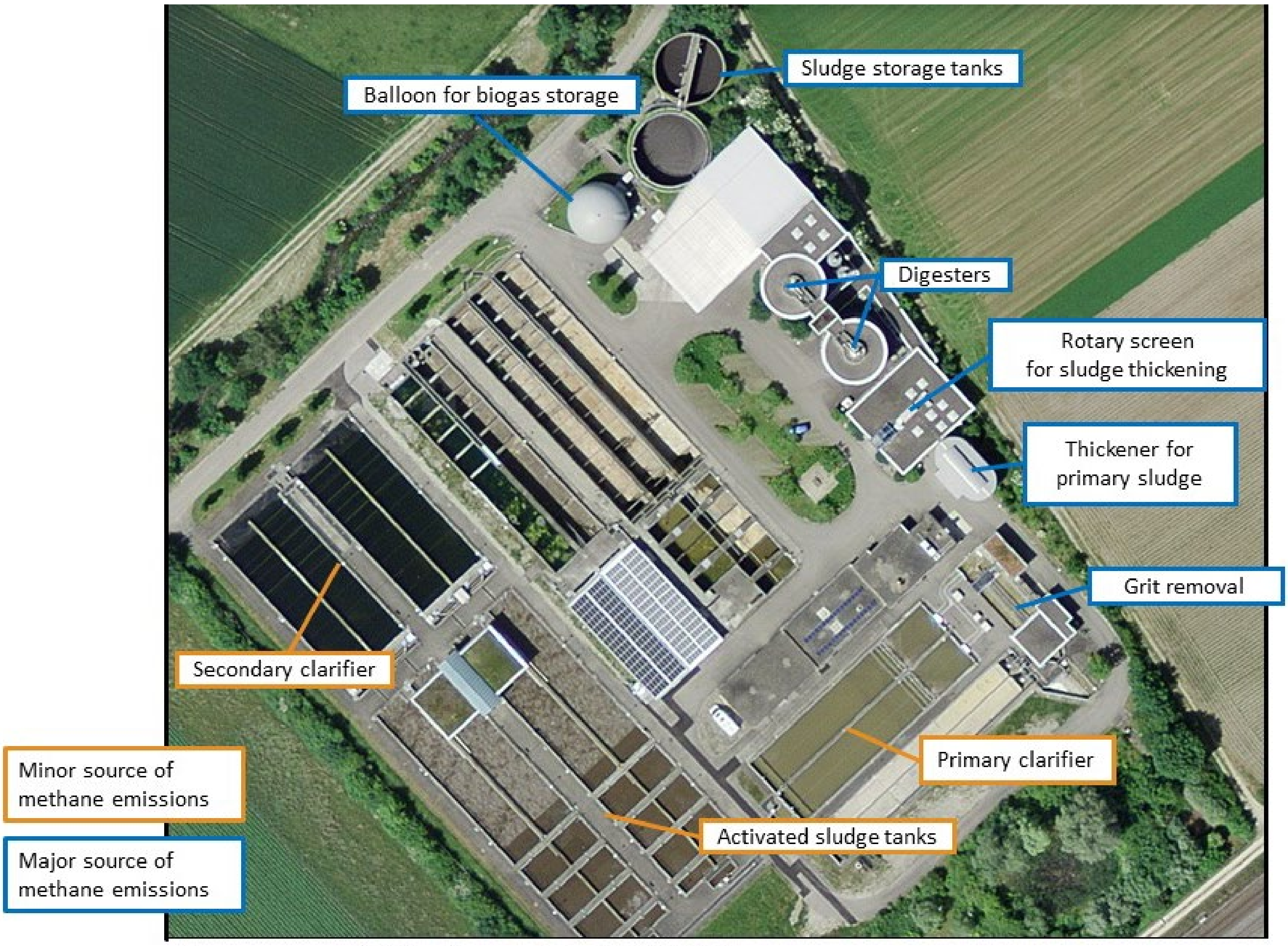

Figure 2: WWTP Moossee-Urtenenbach: overview on the wastewater and sludge treatment units.

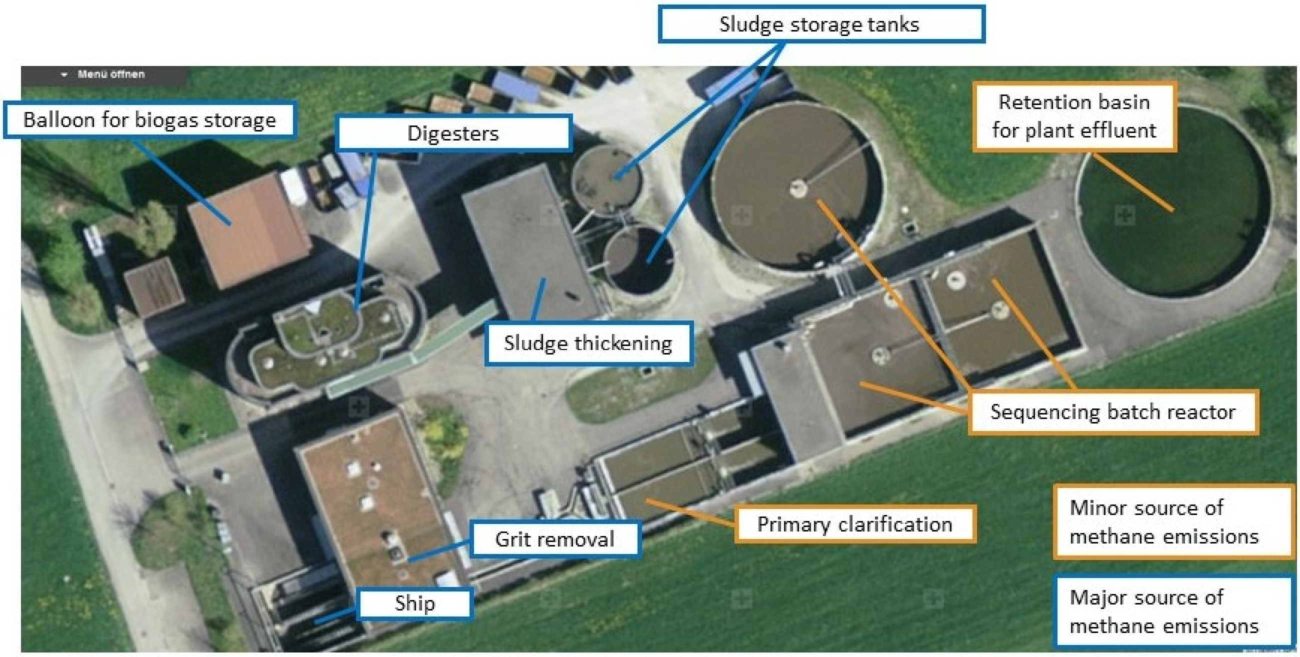

Figure 3: WWTP Gürbetal: overview on the wastewater and sludge treatment units.

After a residence time of approx. 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 the sludge storage tanks (volume: 1000 m^3 each). The sludge is regularly evacuated and transported to the WWTP Bern 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 grit chamber, and within the sludge line the storage tanks, the digesters (if leakages occur), the gas storage, 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 primary sludge enters the digesters with a dry matter content of 2%. The anaerobic digestion is operated at mesophilic conditions. After a residence time of approx. 20 days the sludge is again dewatered to 4% of dry matter after addition of a flocculant by means of a rotary screen and then transferred to the sludge storage tanks. The biogas is fed to a gas motor for electrical power production. The heat is used for regulating the temperature in the digester. The sludge is regularly evacuated and transported to the WWTP Bern for further treatment and disposal. The tanks are stirred daily in the morning. The supernatants are removed from the surface in the afternoon and returned to the plant influent. The gas torch is rarely used.

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

Ammonia emissions will be measured at the WWTP Moossee-Urtenenbach. For NH_3, information on emissions from WWTPs is sparse. Data is available from Samuelsson et al. (2018) only. As for methane, their data suggest that NH_3 mainly originates from sludge treatment.

4. Sources of methane in wastewater treatment plants

4.1 Data on WWTPs from the literature

Daelman et al. (2012) measured the emissions from 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 gaseous samples were determined with gas chromatography. In liquids (i.e. wastewater and sludges), the dissolved methane was measured with the salting-out method.

72% of the total methane emissions came from the processes that are related to the anaerobic digestion: 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.

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. They identified sludge treatment and energy production units as the main CH_4 emission sources but did not provide numbers for individual sources. The total average and median emissions were 0.6 and 0.3% of the CH_4 produced (range: 0.04 to 2.1% of the CH_4 produced). Yoshida et al. (2014) presented similar results from a Danish WWTP. Samuelsson et al. (2018) who used the same method for the emission measurement as Delre et al. (2017) 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 settlers: 4%, activated; sludge reactors: 5%, secondary settlers: 2%)

4.2 Data on emissions from slurry storage tanks from the literature

It can be concluded from the literature that the sludge treatment is the main source of methane emissions from WWTPs. Within the sludge line, it seems likely that the digested liquid sewage sludge stored in open tanks as widespread in Switzerland represents the principal methane source. Since data from the literature on methane is sparse or rather inexistent, data from the agricultural sector could be considered. Table 1 and Table 2 provide emission data from storage of anaerobically digested slurry and untreated, raw cattle and pig slurry from the literature.

Table 1: Emissions of methane from storage of anaerobically digested slurry (references are listed in Appendix 2)

	Environmental c.	Pilot-scale	Lab-scale
n	1	8	13
	$g\ CH_4\ m^{-3}\ h^{-1}$		
Average	0.79	0.15	0.12
Median	0.79	0.07	0.005
Minimum	0.79	0.00	0.000
Maximum	0.79	0.48	0.42

Table 2: Emissions of methane from storage of untreated cattle and pig slurry (references are listed in Appendix 3)

	Environmental c.	Pilot-scale	Lab-scale	Environmental c.	Pilot-scale	Lab-scale
	Untreated cattle slurry			Untreated pig slurry		
n	5	51	14	11	13	16
	$g\ CH_4\ m^{-3}\ h^{-1}$					
Average	0.90	0.75	10.9	1.53	1.14	7.84
Median	0.75	0.44	0.78	0.48	0.97	2.85
Minimum	0.00	0.01	0.00	0.02	0.01	0.01
Maximum	1.94	7.72	51.00	5.01	3.37	33.1

It can be hypothesized that anaerobically digested slurry is a matrix similar to sewage sludge while untreated cattle and pig slurry differs in terms of volatile solids (VS). However, the slurries investigated in the studies providing results given in Table 1 and Table 2 had VS contents in the range between 60% and 80% of dry matter. Untreated slurries exhibit only slightly higher VS contents than anaerobically digested slurries (data not shown). These VS contents are substantially higher than VS in sewage sludge with anaerobic treatment which is around 45% dry matter (Gujer, 1999; Kupper, Chassot, 1999).

For methane emissions, two factors can be relevant: (1) the amount of degradable organic matter, i.e. methane emission from a substrate correlate with the amount of degradable organic matter; (2) the methanogenic potential, i.e. methane emissions from a substrate correlate with the inherent methanogenic activity. Factor 1 suggests that methane emissions are higher from untreated slurries as compared to anaerobically digested slurry. Factor 2 would indicate the opposite. It is not clear from the data presented in Table 1 and Table 2 which factor applies or dominates. Moreover, data from the literature are ambiguous. Holly et al. (2017), Rodhe et al. (2015) and Wang et al. (2014) conducted a direct comparison of methane emissions from untreated and anaerobically digested cattle or pig slurry. In two studies, both at laboratory scale (Holly et al., 2017; Wang et al., 2014), the emissions from the anaerobically digested cattle and pig slurry, respectively, were lower as compared to the untreated slurries while the opposite was observed by Rodhe et al. (2015) in a study at pilot scale. Studies at laboratory scale should be critically evaluated regarding their validity (this applies mainly for Wang et al., (2014) who used a debatable analytical method; Hassouna et al., 2013).

Therefore, data from anaerobically digested slurry is considered as equivalent to untreated slurry with respect to the emission potential for methane. It can be assumed that both substantially deviate from anaerobically digested sewage sludge as produced by Swiss WWTPs. Still, these data are used for the emission estimates since more appropriate data bases are lacking.

The data in Table 1 and Table 2 are arranged according to studies carried out under environmental (i.e. real world) conditions and at pilot or laboratory scale. The emission data vary over a large range. In terms of a preliminary approach, the median values should be used for emission estimates. Data from laboratory studies should be omitted since they highly differ from environmental and pilot scale studies which suggests that lab scale studies are not appropriate to provide data for emission estimates. The median of the median values given in Table 1 and Table 2 from environmental and pilot scale studies results in an emission of 0.8 g CH_4 m^{-3} h^{-1}.

4.3 Conclusions for measurements of individual sources at the Swiss WWTPs

Table 3 shows the data for the emission estimate for the methane emissions from the sludge storage tanks. They suggest an important share of the emissions from the storage tank, i.e. in the range of 70% or more. This complies with the source apportionment of emissions within a WWTP as suggested in the literature presented in section 4.1. It can thus be concluded that an additional measurement of emissions from sludge storage tanks would generate emission data from the prevailing source at WWTPs such as Moossee-Urtenenbach and Gürbetal and that such measurements would be useful to crosscheck the emission data based on an inverse-dispersion technique. Considerations on how such measurements could be realized are provided in the Appendix 4.

Table 3: Data for the emission estimates for the sewage sludge storage tanks of the WWTPs Moossee-Urtenenbach and Gürbetal

	Operating data WWTP		CH_4 emissions		Share of total from storage tanks
	Population equivalents	Average sludge volume in storage tanks	Storage tanks*	Total WWTP**	
	n	m^3	kg CH_4 a^{-1}		%
Moossee-Urtenenbach	35'000	1'400	9'811	14'000	70%
Gürbetal	15'000	800	5'606	6'000	93%

* Basis for the emission estimate: 0.8 g CH_4 m^{-3} h^{-1} based on data from Table 1 and Table 2

** Basis for the emission estimate: 400 g CH_4 PE^{-1} a^{-1} based on Delre et al. (2017)

5. Sources of ammonia in wastewater treatment plant

As performed in the sections 4.1 and 4.2 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} a^{-1} can be estimated for the whole WWTP. For the WWTPs Moossee-Urtenenbach and Gürbetal, this results in annual total emissions of 152 kg and 65 kg NH_3, respectively. 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.

NH_3 emission from the storage of sewage sludge can be estimated based on data from the agricultural sector, i.e. emissions from slurry storage. Based on Kupper et al. (2017) an emission factor for slurry of 0.05 g NH_3 m^{-2} h^{-1} could be applied for a preliminary emission estimate. Based on this data and the surface of the storage tanks, the share of the storage tanks is 83% and 85% for the WWTPs Moossee-Urtenenbach and Gürbetal, respectively.

6. Representativeness of the WWTPs selected for emission measurements and preliminary suggestions for emission modeling within UNFCCC

A comparison of the information provided in section 2 and 3, suggests that the WWTPs as selected can be considered as representative for Swiss WWTPs. A restriction is the fact that both plants transport the final sludge to a large WWTP in the region 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 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 4). A rough estimate could be possible by asking the dimensions of the outdoor stockpiles of the WWTP studied by Samuelsson et al. (2018) and based on this, derive a surface or volume specific emission factor and extrapolate it to conditions common at Swiss WWTPs, or as in this case, the WWTP Bern-Neubrück.

For emission modeling within UNFCCC, it is recommended to use the emission factors generated within the planned measurements. To extrapolate the emission data to the Swiss level (emission per unit activity data), simply available activity data can be suggested such as PE, kg Chemical Oxygen Demand (COD) or kg methane production.

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Appendix 1

Aerial pictures of WWTPs with the surrounding area

Figure 4: WWTP Moossee-Urtenenbach and its surrounding area. Prevailing wind direction: west, south-west. The red and orange lines indicate the approximate position of the line integrated measurement devices.

Figure 5: WWTP Gürbetal and its surrounding area. Prevailing wind direction: north, north-east. The red and orange lines indicate the approximate position of the line integrated measurement devices.

Appendix 2

Emission data from stored anaerobically digested slurry

Studies at environmental conditions

Balde, H., VanderZaag, A.C., Burtt, S.D., Wagner-Riddle, C., Crolla, A., Desjardins, R.L., MacDonald, D.J. 2016. Methane emissions from digestate at an agricultural biogas plant. Bioresource Technol. 216: 914-922.

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Appendix 3

Emission data from stored untreated cattle and pig slurry

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Appendix 4

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.