

# Elaboration of a data basis on greenhouse gas emissions from wastewater management - Final report N2OklimARA

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Authors: Wenzel Gruber<sup>1</sup>, Adriano Joss<sup>1</sup>, Manuel Luck<sup>1</sup>, Thomas Kupper<sup>2</sup>, Marcel Bühler<sup>2</sup>, Tobias Bühner<sup>1</sup>

## Affiliations:

<sup>1</sup>Eawag, Swiss Federal Institute for Aquatic Science and Technology, 8600 Duebendorf, Switzerland

<sup>2</sup>Bern University of Applied Sciences School of Agricultural, Forest and Food Sciences, 3052 Zollikofen, Switzerland

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Eidgenössische Anstalt für Wasserversorgung, Abwasserreinigung und Gewässerschutz, Eawag, Überlandstrasse 133, 8600 Dübendorf

Authors:

Wenzel Gruber, Adriano Joss, Manuel Luck, Thomas Kupper, Tobias Bühler

FOEN support:

Michael Bock, Adrian Schilt, Daiana Leuenberger, Rainer Kegel, Regine Röthlisberger

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Floating flux chamber placed on an activated sludge reactor (M. Luck, Eawag)

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## Summary

The currently applied approaches to estimate direct greenhouse gas (GHG) emissions of nitrous oxide ( $\text{N}_2\text{O}$ ) and methane ( $\text{CH}_4$ ) from wastewater treatment (sector 5D) in the national greenhouse gas inventories of Switzerland are based on the IPCC guidelines (IPCC 2006). A review of the Swiss application of guidelines showed that the guidelines are only partly representative for Switzerland (Eawag 2018). Hence, an extended study was performed including extensive monitoring campaigns on Swiss wastewater treatment plants (WWTPs), analysis of existing data sets and literature reviews. The present document summarizes the study and suggests an improved methodology for an assessment of the GHG emissions of sector 5D.

$\text{N}_2\text{O}$  emission are drastically underestimated with the current methodology. Additionally, source allocation of emissions is not representative. Hence, we suggest a new methodology based on 17 (14 Swiss and 3 from literature) long-term monitoring campaigns on full-scale WWTPs and an analysis of nitrogen flows in Swiss WWTPs. The decreasing  $\text{N}_2\text{O}$  emissions since 1990 link to improvements of nitrogen removal rates on WWTPs. Further reductions are expected in the coming decades because of further planned improvements of nitrogen removal on Swiss WWTPs. Major uncertainties within the new methodology can be linked to the emission factor of WWTPs with carbon removal only. The occurrence of such plants decreased substantially between 1990 and 2020.

$\text{CH}_4$  emissions are overestimated with the current methodology. Additionally, source allocation of emissions is not representative. Hence, we propose a new methodology based on monitoring campaigns and a literature review. Emissions increased moderately since 1990 along with an increase in population.

## 1. Introduction

Wastewater treatment in Switzerland is generally performed in centralized wastewater treatment plants (WWTPs) fed by closed sewer systems. This is reflected by the high connection rate (97.3% in 2018) of the population to centralized WWTPs (FOEN (2020b)). Primary, physical treatment and secondary, biological treatment for the removal of carbon compounds are performed at all Swiss WWTPs (FOEN 2021a). Tertiary, biological treatment is common in Switzerland and involves various biological processes to remove nutrients such as nitrogen, and phosphorus. The design of the biological process (secondary and tertiary treatment) depends, among other factors, on the nutrient removal goal, which covers, at least, carbon (secondary treatment) and phosphorus (tertiary treatment) removal. Depending on the discharge requirements, nitrogen transformation (to nitrate) or full nitrogen removal is implemented (Gujer 2007). Secondary and tertiary treatment lead to the production of sludge, which is typically treated and stabilized in anaerobic digestion processes. The digested sludge is incinerated and subsequently landfilled. Quaternary treatment of wastewater to remove micropollutants is gradually implemented in Switzerland.

Onsite emissions of greenhouse gases (GHG), typically methane ( $\text{CH}_4$ ) and nitrous oxide ( $\text{N}_2\text{O}$ ), occur in parts of the wastewater treatment process. Tertiary treatment of wastewater can cause onsite emissions of  $\text{N}_2\text{O}$  during biological nitrogen removal. These emissions were reported to exhibit a strong seasonal pattern in some cases (Vasilaki et al. 2019). Hence, long-term monitoring campaigns are required to assess representative emissions factors (EFs) (Daelman et al. 2013). Similarly,  $\text{N}_2\text{O}$  is produced during transformation of nitrogen from the discharged water in the environment and the subsequent emissions are accounted to the Waste sector (IPCC 2006). The sewer system and the anaerobic digestion of sludge can lead to emissions of  $\text{CH}_4$  through leakage. Energy demand for the operation of WWTPs causes related  $\text{CO}_2$  emissions, which are not accounted to the sector Waste according to the 2006 IPCC guidelines (GL).

Switzerland applies the 2006 IPCC guidelines to calculate the GHG emissions from the wastewater sector in the national inventory report (NIR). The expert review (ER) by the authors of this study (Eawag 2018 ) showed that the calculations and reporting are mostly carried out correctly and following the guidelines (IPCC 2006). However, the review also showed that the IPCC 2006 GL and the corresponding Swiss application (FOEN 2017) are only partly representative to account for the emissions from wastewater treatment in Switzerland. Three main issues could be characterized: (i) The nitrogen mass balance applied for WWTPs does not include nitrogen removal through denitrification. (ii) The emission factor for N<sub>2</sub>O from biological treatment is drastically lower than values reported in representative scientific publications. (iii) Activity data for CH<sub>4</sub> emissions are based on an uncommonly used unit (kgBOD/Person/year). With the Refinements of the GL in 2019 (IPCC 2019b) the issues could only partly be resolved (Gruber et al. 2021). Hence, we recommend that a country-specific methodology is used for Switzerland.

This report presents the evaluation of current and historic performance and emission data from WWTPs and, as a result, proposes new methodologies to estimate GHG emissions from wastewater treatment (N<sub>2</sub>O, CH<sub>4</sub>). In particular, activity data (AD) and emission factor (EF) estimation methods are evaluated and verified with representative data for N<sub>2</sub>O and CH<sub>4</sub> emissions. The proposed methodologies are finally compared with the current approaches applied for the Swiss NIR. This work is based on a 4-years research project yielding several publications, emissions monitoring guidelines and two master theses (Bührer 2020, Luck 2018).

## 2. Activity data (AD)

### 2.1. N<sub>2</sub>O AD: Nitrogen loads to wastewater treatment

#### 2.1.1. Assessment of the reporting according to 2006 IPCC guidelines (Expert review)

In the NIR of Switzerland (FOEN 2020a), the total nitrogen loads in the influent of WWTPs are calculated based on countrywide protein consumption, as described the 2006 IPCC guidelines (GL, IPCC (2006)). In the corresponding mass flow model, nitrogen removal in WWTPs is only possible via incorporation of nitrogen in the sludge (Figure 1). Denitrification is not considered in the GL although it typically represents the most substantial nitrogen removing process in Swiss WWTPs. Hence, the application of the 2006 IPCC guidelines for the estimation of nitrogen loads in the inflows and outflows of WWTPs was considered as not representative for Switzerland in our ER (Eawag 2018). Total nitrogen loads extracted from plant specific performance data sets for Switzerland were suggested as an alternative for the estimation (Eawag 2018, Strähl et al. 2013).

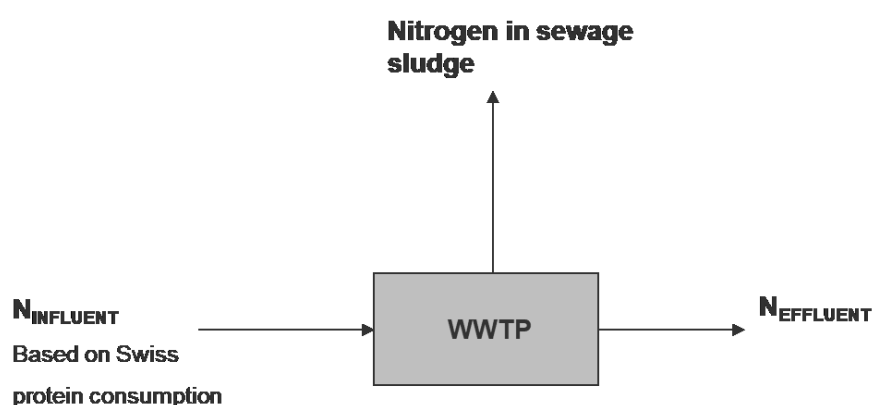


Figure 1 Nitrogen flow model in WWTPs used in the NIR 2020

In the 2019 refinement of the guidelines, nitrogen removal via denitrification is implemented for the estimation of the effluent loads (IPCC 2019b). However, nitrogen inflow loads are still estimated using the protein consumption, as in the 2006 IPCC guidelines (IPCC 2006). Hence, we suggest a method for the estimation of nitrogen loads in WWTPs, given the good data

availability for Switzerland. In the following, we evaluate and compare the total nitrogen loads extracted from several data sources.

### 2.1.2. Data analysis

The yearly, total and relative (per inhabitant) nitrogen loads in WWTPs were calculated or extracted from the available data sets and reports (Table 1): data sets by Strähl et al. (2013) and FOEN (2021a) contain nitrogen load data for roughly 70 and 75 %, respectively, of the Swiss population. Reports by BUWAL (1993), (FOEN 2006), FOEN (2010), and FOEN (2013) summarize estimations of the total nitrogen flows in WWTPs as a part of the overall nitrogen balance of Switzerland.

Table 1 Estimated nitrogen loads, inhabitants and nitrogen removal rate for reference years according to representative data sources

Reference	Year	N <sub>INFLUENT</sub> load [tN/year]	Inhabitants	N <sub>INFLUENT</sub> load [gN/P/day]	N <sub>EFFLUENT</sub> load [t/year]	N Removal rate (%)
BUWAL (1993)	1990	47'000	6,673,000	19.3	42,000	11
(FOEN 2006)	1996	46'000	7,020,000	18.0	32,200	30
FOEN (2010)	2005	43,200	7,415,000	16.0	26,000	40
(Strähl et al. 2013)	2010	46,490	7,785,806	16.4	23,830	48
FOEN (2021a)	2020	45,794	8,606,000	14.6	21,276	53
IPCC (2006)	2015	65,780	8,282,000	21.8	47,550	

To compare the different data sets, we calculated the relative influent nitrogen loads per year and inhabitant. The results vary substantially (Table 1). The values for 1990 are substantially higher than for the later data sets. The total nitrogen loads are clearly lower for all references than the applied value in the NIR 2020 based on the 2006 IPCC guidelines (FOEN 2020a).

The nitrogen removal rates exhibited a strong increase between 1990 and 2010 (Table 1). The value for 2020 suggests a strongly increased nitrogen removal rate, which confirms previous projections of Swiss nitrogen flows by the FOEN made in 2013 (FOEN 2013). The removal rates reflect the increase of WWTPs with full nitrogen removal as a treatment goal in



Switzerland until 2010. Similar values are proposed in the 2019 update of the guidelines (IPCC 2019b). However, applying the 2019 default values would still lead to an overestimation of nitrogen removal for the case of Switzerland.

### 2.1.3. Proposed methodology

Based on the evaluation of nitrogen flows in Swiss WWTPs, we suggest a methodology to estimate nitrogen loads as activity data for N<sub>2</sub>O emissions from WWTPs in Switzerland (Figure 2). We propose to estimate the total nitrogen in the influent based on a fixed value of nitrogen released per inhabitant and year. For the nitrogen removal rate we suggest to interpolate available data sources. Other variables are either computed or estimated as in the NIR from 2020 (FOEN 2020a).

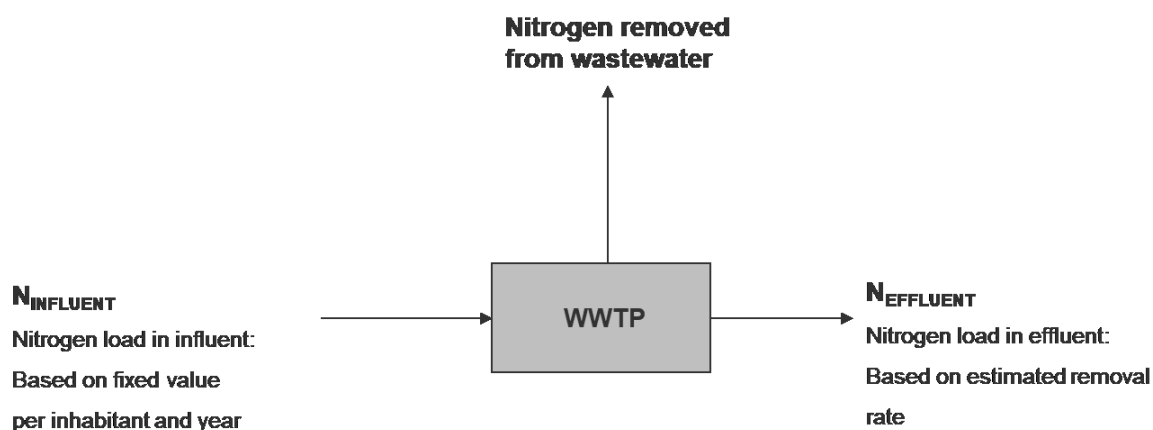


Figure 2 Proposed nitrogen flow model in Swiss WWTPs

The nitrogen mass flows are computed using equations 1 and 2:

Influent nitrogen load for the calculation of onsite N<sub>2</sub>O production:

$$N_{INFLUENT} = P * N * T_{PLANT} \quad (1)$$

Effluent nitrogen load based on the average removal rate – substituting Equation 2 from the IPCC 2006 Guidelines:

$$N_{EFFLUENT} = N_{INFLUENT} * (1 - r_{NITROGEN\ REMOVAL\ RATE}) \quad (2)$$

Output variables:

$N_{INFLUENT}$	Influent nitrogen load [kg N / year]
$N_{EFFLUENT}$	Effluent nitrogen load [kg N / year]

Input variables:

$P$	Population [Persons]
$T_{PLANT}$	Connection rate to WWTPs [-]
$N$	Nitrogen load [kg N / Person/ year]
$r_{NITROGEN\ REMOVAL\ RATE}$	Average nitrogen removal rate [-]

The estimation of single variables for past and future estimations as well as the estimation of corresponding uncertainties is provided in the following.

### **Population ( $P$ )**

The population size is a standard value required for the NIR. Uncertainty is not considered for this variable.

### **Connection rate to WWTPs ( $T_{PLANT}$ )**

The connection rate to WWTPs is a standard value required for the NIR. Uncertainty is not considered for this variable. The values was stable at 97.3 % since 2011 (FOEN 2021b) as displayed in the following figure.

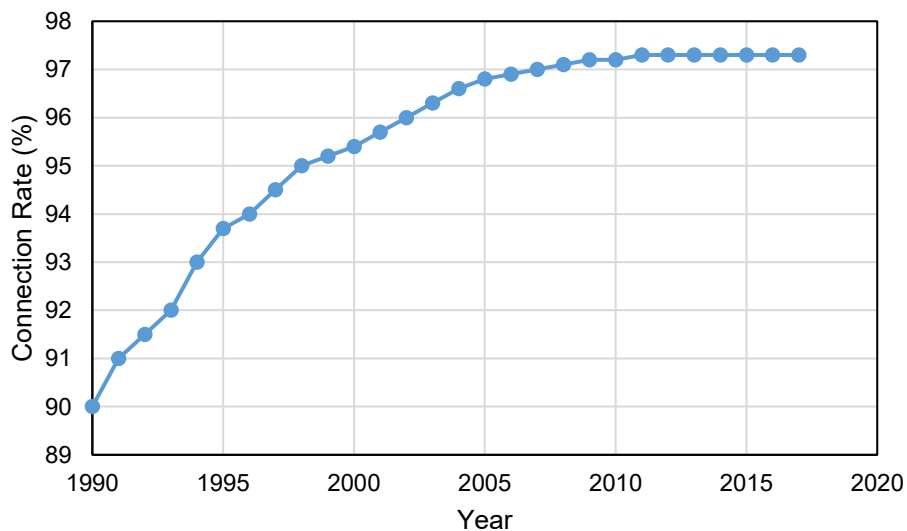


Figure 3 Connection rate to centralized wastewater treatment since 1990 (FOEN 2021b)

### **Nitrogen load ( $N$ )**

We estimate the nitrogen load per inhabitant and year by interpolating the available data sources (Table 1). The decreasing nitrogen load per inhabitant can be explained by the decreasing production of pork meat and beef in Switzerland since 1990 (Baur and Kraye 2021). Consequently, the amount of industrial protein discharged to the wastewater was strongly reduced. The standard error of the linear interpolation is equal to 0.5 gN/P/d. Consequently, the uncertainty equals to 1.5 kgN/P/y ( $0.5 \text{ gN/P/D} \times 2.776$  (=coverage factor: student's  $t$  for 4 degrees of freedom)) or 10% of the average value.

The proposed nitrogen load is higher compared to other references, which typically project a value of 14 gN/inhabitant/day (Larsen and Gujer 1996). Most likely, this can be explained by nitrogen discharged by industry. To reduce uncertainties in future estimations, the value should be updated with follow-up data sets of the reference FOEN 2020a, which are expected until 2025 according to the FOEN section 'Water Body Protection' in the Water division.

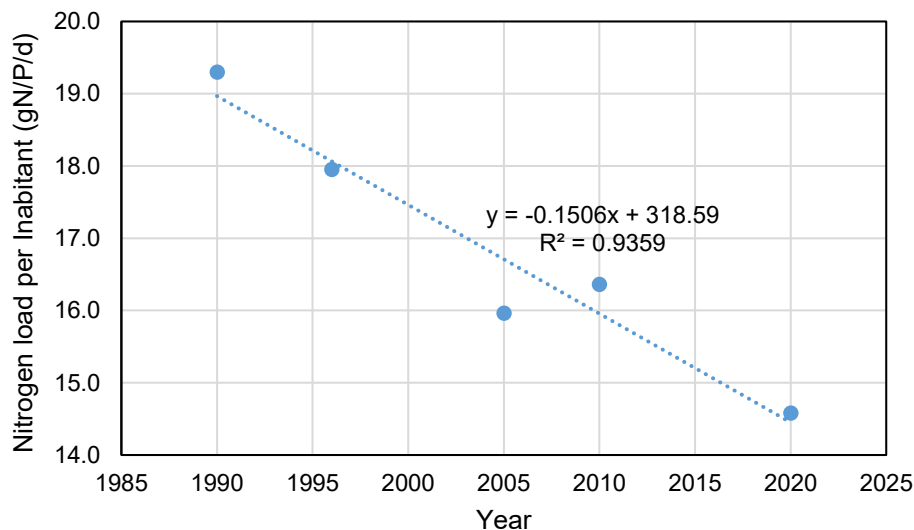


Figure 4 Nitrogen load in wastewater per inhabitant and day

#### **Average nitrogen removal rate ( $r_{\text{NITROGEN REMOVAL RATE}}$ )**

Nitrogen removal rates are calculated by linear interpolation of the available data points (Table 1, Figure 5). For this application all five data sources are included. The different growth of the removal rates between 1990 and 1996, 1996 and 2010, and after 2010 is modeled using three separate linear functions. A further moderate increase until 2025 can be expected due to the tendency to target higher nitrogen removal rates if a WWTP is refurbished. After 2025 a strong increase of the removal rate is expected due to the political decision to substantially increase nitrogen removal in WWTPs (Swiss Parliament 2021). For future estimations, the average value should be updated with follow-up data sets of the reference FOEN 2020a, which are expected until 2025 according to FOEN section 'Water Body Protection'.

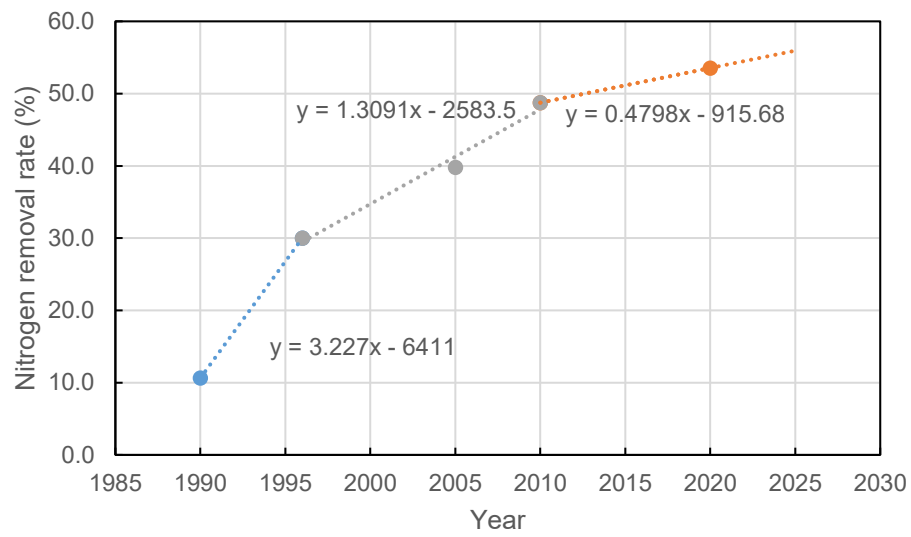


Figure 5 Nitrogen removal rates for years with reference data and interpolation functions.

## 2.2. CH<sub>4</sub> AD: Carbon loads to wastewater treatment and gas production

### 2.2.1. Assessment of reporting according to 2006 IPCC guidelines (Expert Review)

In the NIR of Switzerland, the loads of organic carbon in the influent of WWTPs are calculated based on the biological oxygen demand (BOD). This is not ideal because BOD is no longer measured at Swiss WWTPs and, consequently, the comparison of values with performance data of WWTPs is difficult. Hence, it was suggested to change to the chemical oxygen demand (COD) as a unit with a conversion factor of 2 kgCOD/kgBOD and rename the variable from BOD to COD (Eawag 2018). Additionally, changes in the accounting of sewage gas from WWTPs were proposed. Firstly, sewage gas usage in the Federal statistics (SFOE 2017) only reports part of the total production and ignores leakage from sludge treatment. The emissions during sewage sludge treatment were not accounted at all. Secondly, gas losses during transformation into usable energy was double counted and should not be accounted to the Waste sector. In the 2019 refinement of the guidelines, no major changes were proposed for the assessment of carbon loads to WWTPs (IPCC 2019).

### 2.2.2. Proposed methodology

To calculate the AD for CH<sub>4</sub> emissions (equations 3-4), we propose the same methodology as in the ER (Eawag 2018). For the full calculation scheme see chp. 4.2.

Total organically degradable material:

$$TOW_{SEWER} = P * T_{SEWER} * COD_{pd} * 0.001 * I * 365 \quad (3)$$

Sewage gas usage reported in Federal statistics:

$$P_{REP, SEWAGE GAS} = \sum P_{j, SEWAGE GAS} \quad (4)$$

Output variables:

$TOW_{SEWER}$	Organically degradable material in sewer [kg COD / year]
$P_{REP, SEWAGE GAS}$	Sewage gas usage reported in Federal statistics
$P_{TOT, SEWAGE GAS}$	Total gas production including unaccounted pathways

Input variables:

P	Population [# person]
$T_{SEWER}$	Connection rate to sewer system = connection rate to WWTPs [-]
I	Correction factor for additional industrial COD discharged into sewers [-]
$COD_{pd}$	Per capita load of organically degradable material [g COD / pers. / day]
$P_j$	Reported sewage gas usages in Federal statistics [TJ / year]

The estimation of single variables for past and future estimations as well as the estimation of corresponding uncertainties is provided in the following.

### ***Population (P)***

The population size is a standard value required for the NIR. Uncertainty is not considered for this variable.

### ***Connection rate to WWTPs ( $T_{PLANT}$ )***

The connection rate to WWTPs is a standard value required for the NIR. Uncertainty is not considered for this variable. The values was stable at 97.3 % since 2011 (FOEN 2021b).

### ***Correction factor for additional industrial COD discharged into sewers (I)***

The correction factor for additional industrial COD discharged into sewers is a standard value required for the NIR (1.25; IPCC 2006 default value). Uncertainty is not considered for this variable. The default value according to IPCC (2006) can be applied.

### ***Per capita load of organically degradable material ( $COD_{pd}$ )***

The per capita load of organically degradable material is defined as 120 gCOD/P/d in the ER (Eawag 2018). This value is commonly used in standard wastewater treatment text books applicable to Switzerland (Gujer 2007). Uncertainty is not considered for this variable.

***Reported usages in Federal statistics (P<sub>i</sub>)***

The usage of sewage gas is estimated as in previous versions of the NIR (FOEN 2020a) based on the federal statistics (SFOE 2017). Only the emissions caused by leakage, sewage gas upgrade and combustion in torches are accounted to the waste sector. Uncertainties are not considered for these variables.



### **3. Emission factors (EF)**

#### **3.1. N<sub>2</sub>O EF**

##### **3.1.1. Assessment of 2006 IPCC guidelines (Expert Review)**

In the NIR of Switzerland, the N<sub>2</sub>O EF for WWTPs is based on the default value of the 2006 IPCC guidelines (IPCC 2006). The value applied (3.2 gN<sub>2</sub>O/inhabitant/year) was shown to drastically underestimate onsite emissions from WWTPs (Eawag 2018). However, data availability was insufficient to make robust predictions about the EF in 2018, since most of the monitoring campaigns published are based on short-term monitoring campaigns of a few days to months (Eawag 2018). Long-term monitoring campaigns of at least one year are required to representatively estimate EFs from WWTPs due to the substantial seasonal variation of N<sub>2</sub>O emissions (Daelman et al. 2013, Gruber et al. 2020). In our ER, a new emission model was introduced to better represent the emission process in WWTPs. The EF for N<sub>2</sub>O produced through the discharge of nitrogen into the environment was considered to be representative (Eawag 2018).

The EF was substantially increased from 0.035% in the 2006 IPCC guidelines (Czepiel et al. 1995, Daelman et al. 2015, IPCC 2006) to 1.6% in the 2019 Refinement to the 2006 IPCC guidelines (IPCC 2019a). However, also the latter is an average value of mostly short-term monitoring campaigns and therefore not representative as our analysis shows (Gruber et al. 2021). Due to the lack of long-term monitoring campaigns, an extensive monitoring campaign was conducted including long-term (> 1 year) data of 14 WWTPs. In the following, we describe the data assessed and deduce a method to estimate countrywide emissions for Switzerland.

##### **3.1.2. Monitoring and data analysis**

###### ***Selection of WWTPs***

WWTPs were selected to represent the range of WWTPs in Switzerland. It was assumed that the nutrient removal goal of the biological treatment and the process for the biological process are the most important factors to govern the N<sub>2</sub>O EF from WWTP. Carbon removal only,

ammonium oxidation and full nitrogen removal are removal goals typically found in Switzerland. Biological treatment processes include different configurations of activated sludge and biofilm systems. The resulting selection of WWTPs is shown in Table 2. Additionally, we included the results from three long-term monitoring campaigns published (Chen et al. 2019, Daelman et al. 2015, Kosonen et al. 2016).

Table 2 Selection of WWTPs for the monitoring study according to the process for biological treatment and the nutrient removal goal. Countries: CH, Switzerland; DK, Denmark; NL, the Netherlands; FI, Finland. Processes: A/I, alternately fed and intermittently aerated activated sludge treatment; A2O, anaerobic, anoxic and aerobic cascaded activated sludge treatment; AO, anoxic and oxic cascaded activated sludge treatment; CARR, carousel activated sludge treatment; CAS, conventional activated sludge; FB, fixed bed biofilm reactor; IFAS, integrated fixed film activated sludge; SBR, sequencing batch reactor.

Process	C-Elimination	Nitrification	Denitrification
Activated sludge (plug flow; CAS)	<b>Giubiasco (CH)</b> <b>C-Elimination #2 (CH)</b>		
Activated sludge (plug flow; AO, A2O)		<b>Altenrhein (CH)</b> <b>Viikinmäki (FI)</b>	<b>Schönau (CH)</b> <b>Moossee (CH)</b> <b>Hofen</b>
Activated sludge (SBR)		<b>Uster (CH)</b>	<b>Birs (CH)</b>
Activated sludge (A/I)			<b>Luzern</b> <b>Werdhölzli</b>
Activated sludge (CARR)			<b>Kralingseveer (NL)</b> <b>Avedore (DK)</b>
Hybrid (IFAS)		<b>Langmatt</b> <b>Bazenheid</b>	
Fixed bed		<b>Altenrhein</b> <b>Bern</b>	

### **Off-gas monitoring system**

To monitor the emissions, an existing approach by Chandran et al. (2016) using flux chambers for off-gas collection on full-scale WWTPs was adapted to operate as an online monitoring system for concentrations of N<sub>2</sub>O in the off-gas of WWTPs (Figure 6). Long-term stability and high-frequency measurements were found to be the most important factors of a monitoring system, due to the seasonal and diurnal variability of emissions. Monitoring strategies were developed for different wastewater treatment types (Gruber et al. 2020). Emissions were

calculated by multiplying concentrations of  $N_2O$  in the off-gas and the air flows in the biological reactors (Gruber et al. 2021). Emissions from unaerated zones in the biological treatment were not monitored (Figure 6). Previous research suggests an emission share between 8 and 25% from unaerated zones (Chen et al. 2019, Mikola et al. 2014). The full description of the monitoring system and the calculation methods can be found in the Supporting Information of our publication Gruber et al. (2021).

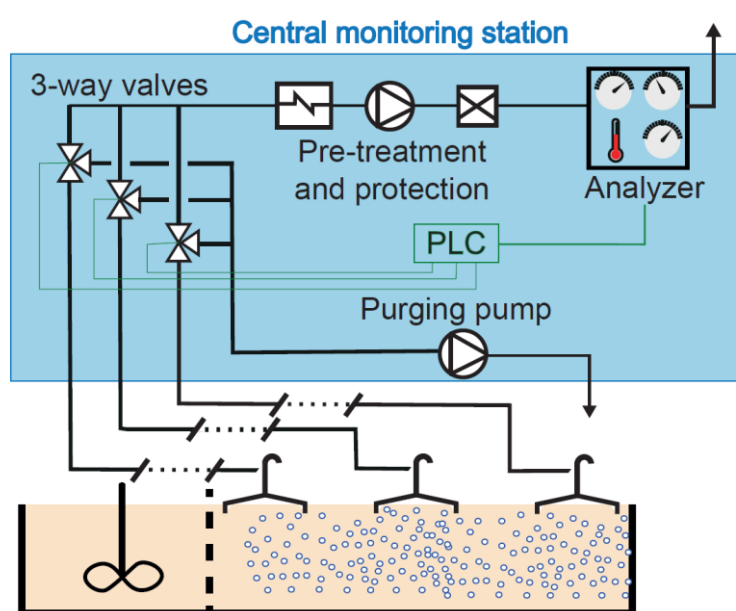


Figure 6 Monitoring system applied for off-gas monitoring on biological wastewater treatment

### ***Emission factors assessed and key factors***

The 14 monitoring campaigns were evaluated in terms of yearly emissions and an average EF was calculated for each WWTP. The EFs assessed were compared with available literature data (Chen et al. 2019, Daelman et al. 2015, Kosonen et al. 2016). In general, the EFs are highly variable (Figure 7) emphasizing the need for an improved method to assess EFs from WWTPs. 11 of the 14 monitoring campaigns are further described and evaluated in a peer-reviewed publication (Gruber et al. 2021). The monitoring data can be found in the Supporting Information of our publication Gruber et al. (2021). The 3 unpublished monitoring campaigns conducted by the authors of this study are further described by Gruber et al. (2022). Compared to the published data we reevaluated the nutrient removal category and set a minimal nitrogen

removal rate of 65% and a full year nitrogen removal for the nutrient removal category 'denitrification'. These requirements are equally set in the data set FOEN (2021a), which is the data basis for our estimation approaches.

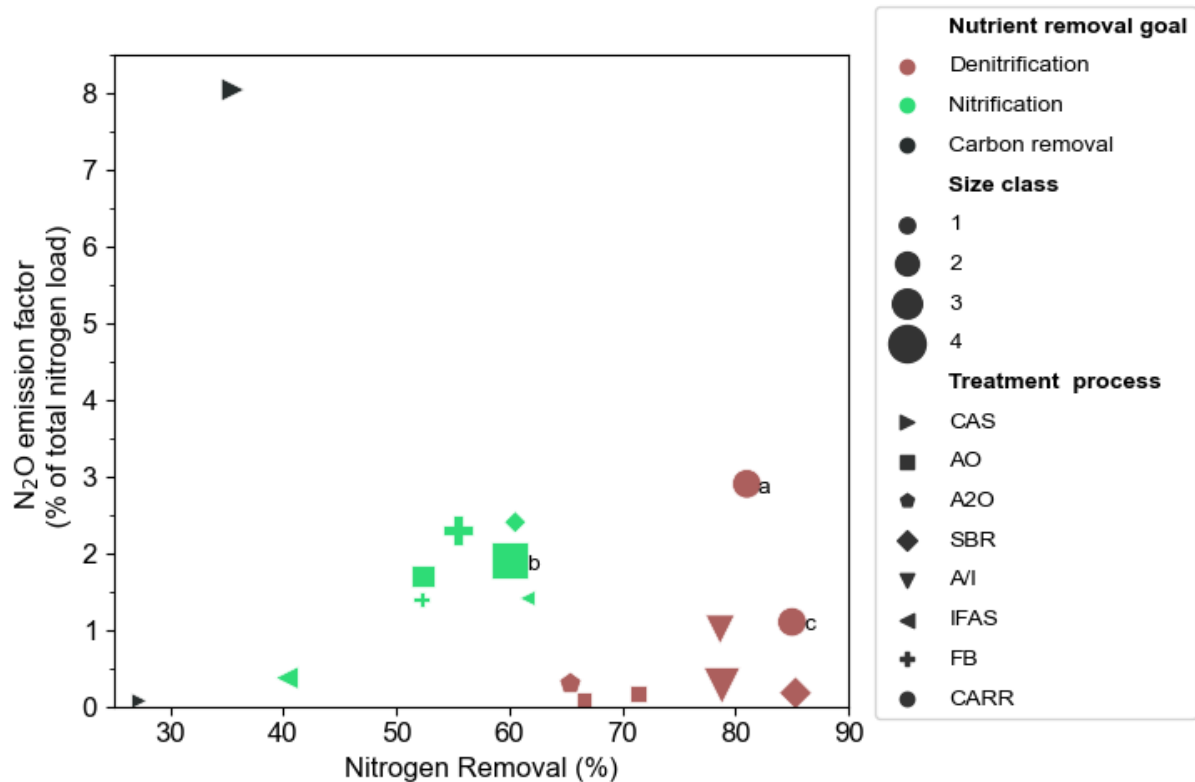


Figure 7 EF assessed for the WWTPs monitored as a function of the nitrogen removal rate of the respective biological process. Small letters highlight studies from literature (a: (Daelman et al. 2015), b: (Kosonen et al. 2016), c: (Chen et al. 2019)). Size of data points describes the size class of the WWTPs (1: < 50,000 PE, 2: 50,000 – 200,000 PE, 3: 200,000 – 500,000 PE, 4: > 500,000 PE). Shape of data points indicates treatment process as described in Table 2.

Correlations of the N<sub>2</sub>O EF with potential drivers were checked to characterize determinants for a countrywide extrapolation. Nitrite concentrations in the effluent and nitrogen removal were found to be the most important factors explaining the N<sub>2</sub>O EF. However, data on nitrite concentrations are very limited on a countrywide level and nitrogen removal alone cannot be linked to the N<sub>2</sub>O EF.

### 3.1.3. Proposed methodology

In our ER, two on-plant emissions pathways were introduced: one for stripped N<sub>2</sub>O during aeration, and one for unstripped N<sub>2</sub>O. To simplify the methodology proposed by our ER (Eawag

2018), we propose to include only two emissions processes (on plant emissions and emissions from effluents) in the emission model (Figure 8).

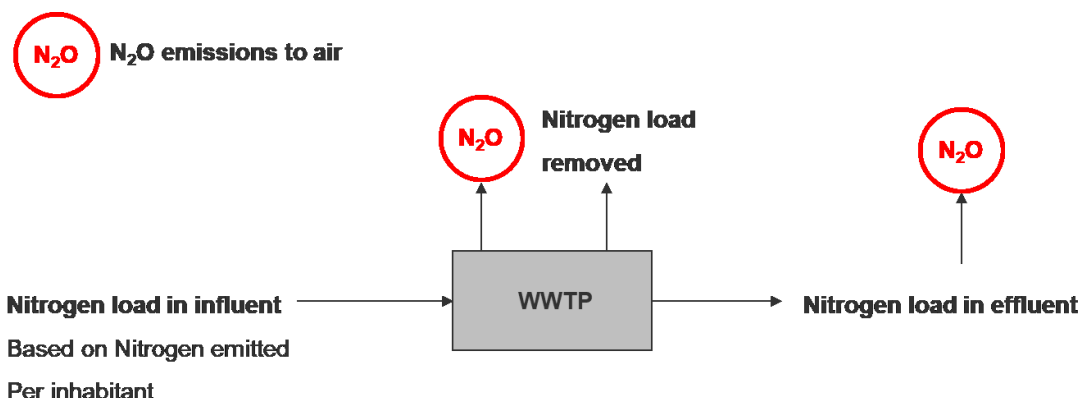


Figure 8 N<sub>2</sub>O emission model for wastewater treatment

We suggest calculating an EF for onsite emissions from WWTPs based on the EFs of the three nutrient removal categories of carbon removal, nitrification only, and year-round nitrogen removal with the overall assumption that a lower nitrogen removal results in higher emissions and a higher probability of nitrite accumulation unless nitrification can be completely excluded. The estimation of the countrywide EF is calculated by multiplying the respective share of nitrogen load treated in WWTPs belonging to a nutrient removal category with the respective EF. We suggest to increase the estimated countrywide EF for the on-plant emissions by a fixed share to incorporate emissions from unaerated zones, since a good correlation between EFs from aerated and unaerated parts has been shown. The EF for N<sub>2</sub>O emitted from nitrogen in the effluent of WWTPs is kept at the default value in the IPCC guideline, as suggested by the ER (Eawag 2018).

The EF is computed using equation 5:

N<sub>2</sub>O EF emissions from WWTP:

$$EF_{N_2O,PLANT} = (1 + F_{N_2O,UNAERATED}) \sum_i EF_{N_2O,PLANT,i} * S_i \quad (5)$$

Output variables:

$EF_{N_2O,PLANT}$  EF for on-plant N<sub>2</sub>O emissions [kg N<sub>2</sub>O-N / kgN]

Input variables:

$F_{N_2O,UNAERATED}$  Share of emissions from unaerated process stages [-]

$EF_{N_2O,PLANT,i}$  EF for removal category type i [kg N<sub>2</sub>O-N / kgN]

$S_i$  Share of total nitrogen treated [-]

The estimation of single variables for past and future estimations as well as the estimation of corresponding uncertainties is provided in the following.

#### ***Share of emissions from unaerated process stages ( $F_{N_2O,UNAERATED}$ )***

Emissions from unaerated parts of a WWTP include emissions of N<sub>2</sub>O produced during aeration and emitted in following process stages (e.g. secondary clarifiers) and the production and emission of N<sub>2</sub>O in anoxic tanks for denitrification. A previous study estimated emissions shares from unaerated processes zones of the biological treatment at 15% (Chen et al. 2019). Another study estimated the share of emissions from secondary clarifiers on average at 19% (Mikola et al. 2014). For unaerated parts, we estimate a share of 25% of the calculated EF from aeration, which is less than the sum of both studies, since not all WWTPs have anoxic zones or secondary clarifiers. We do not expect any time or nutrient removal category dependency of the factor ( $F_{N_2O,UNAERATED}$ ) and therefore assume a constant factor over the whole reporting period.

**Emission factor wastewater treatment ( $EF_{N_2O,PLANT,i}$ )**

We suggest to replace the average values, as suggested by Gruber et al. (2021), by median values and to recalculate the median and standard deviation including all monitoring campaigns in Figure 7. The resulting values are shown in Table 3. The median EFs should be updated if further studies meeting our requirements for monitoring campaigns (at least one year, spatially resolved) are available (Gruber et al. 2020). As long as N<sub>2</sub>O mitigation strategies are not widely implemented on full-scale WWTP, which is not expected, the EFs can be assumed for the whole reporting period since 1990. The EF for the carbon removal WWTPs (category C) exhibits by far the largest standard mean error and uncertainty. Hence, monitoring more carbon removal WWTPs could help to reduce the overall uncertainty. However, the declining importance of carbon removal WWTPs (Figure 9) shows that this mainly improves estimates between 1990 and 2010.

Table 3 Average N<sub>2</sub>O EFs for respective nutrient removal categoriers given in % of the N influent.

	Full year nitrogen removal	All year nitrification	Carbon removal
Median (kg N <sub>2</sub> O-N / kgN)	0.4	1.8	4.3
Standard mean error (kg N <sub>2</sub> O-N / kgN)	0.3	0.3	4.3
Standard mean error relative to median (%)	71	17	98

**Emission factor effluent ( $EF_{N_2O,EFFLUENT}$ )**

The EF for N<sub>2</sub>O (0.5%) emitted from nitrogen in the effluent of WWTPs is kept at the default value in the IPCC guideline (IPCC 2006) due to limited data, as suggested by the ER (Eawag 2018).

### Share of total nitrogen load treated in nutrient removal categories ( $S_i$ )

The share of nitrogen load treated in WWTPs of the three nutrient removal categories for selected reference years (1990, 2010, 2020) was derived using the available data sources (BUWAL 1993, FOEN 2021a, Strähl et al. 2013). Note that data of FOEN (2021) have not yet been validated until publication of this document but a final data set will become available later. The resulting estimations of shares and the respective linear interpolations are shown in Figure 9. For future estimation, we propose that the follow-up data-sets of FOEN (2021a) are used, with expected publication in 2025 according. Until 2025 our projections as in Figure 9 can be applied as no better option is available today. We assume that no further WWTPs with nitrification only are built in Switzerland. The reduced share of carbon removal WWTPs will be compensated by all-year denitrifying plants.

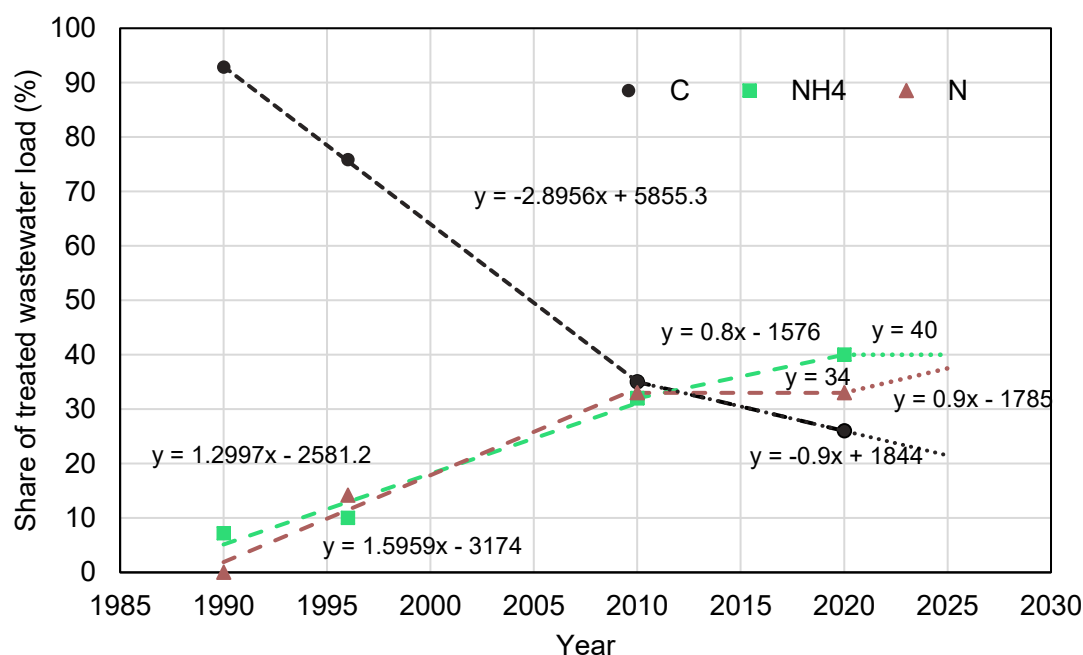


Figure 9 Shares of the treated nitrogen load treated in WWTPs of the three nutrient removal goals (C=carbon removal, NH4 = All-year nitrification, N=full year nitrogen removal) in three reference years (1990, 2010, 2020). The values between the referenced years were assumed by linear interpolation and correlation rules are given in black. The dotted lines represent our projections after 2020.

Concerning on-site emissions, we propose an emission factor called  $EF_{N_2O_{influent}}$  in the accompanying spreadsheet of 2.5% for the year 2019.



## 3.2. CH<sub>4</sub> emission factors

### 3.2.1. Assessment of 2006 IPCC guidelines (Expert Review)

In the current NIR of Switzerland, the CH<sub>4</sub> EFs from WWTPs are based on the default values of the 2006 IPCC guidelines (IPCC 2006) and the Swiss energy statistics for losses from gas treatment (Eawag 2018). Losses during the production of sewage gas and storage of sludge are not included in the 2006 IPCC guidelines (Eawag 2018). Moreover, emissions during conversion of sewage gas to energy (energy line) were found to be double counted in the energy sector and wastewater treatment. Consequently, a new methodology (Figure 10) was proposed in the ER to better represent the emission process on WWTPs (Eawag 2018). To validate the emission model and the EFs, a master thesis was performed to estimate onsite CH<sub>4</sub> from WWTPs based on measurements (Bührer 2020) and a detailed literature review was elaborated in a collaboration with the HAFL (Kupper et al. 2018). Additionally, whole-plant measurements CH<sub>4</sub> emissions were conducted by HAFL (Bühler et al. 2021).

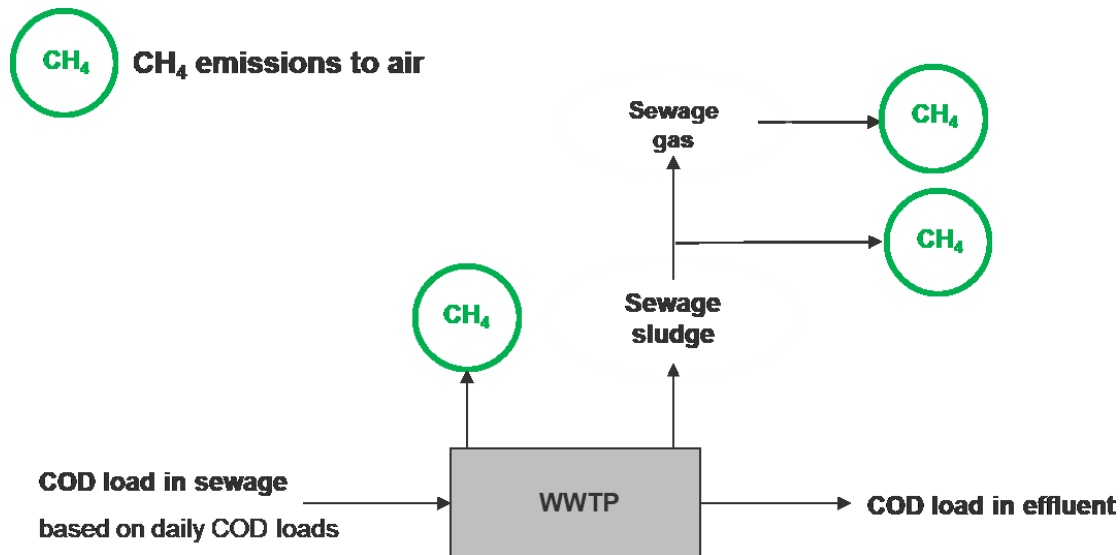


Figure 10 CH<sub>4</sub> emission model for wastewater treatment according to the ER (Eawag 2018)

### **3.2.2. Literature review and monitoring campaigns**

In the whole wastewater treatment process, CH<sub>4</sub> is produced under anaerobic process conditions, typically found in the sewer system and during anaerobic sludge treatment. Emissions can occur in or after the respective process stages (e.g. the water line of the wastewater treatment). For this short review, we evaluated emissions for the three relevant processes: the sewer system, the water line (wastewater treatment), and the sludge line (sludge treatment and sludge storage). For the emissions from WWTPs (water line + sludge line), we compared whole-plant measurements based on the inverse dispersion method or the tracer dispersion method with process stage specific monitoring approaches based on flux chambers (Bührer 2020, Kupper et al. 2018). The energy line of WWTPs was not included in the review, since representative data are available for Switzerland and the respective emissions do not account to the waste sector, but the energy sector.

#### ***Sewer system***

CH<sub>4</sub> production in sewers is complex and can lead to substantial emissions (Fries et al. 2018, Liu et al. 2015b). However, the available studies are barely transferable to Switzerland due to different climatic and geographical conditions. In the ER an EF of 0.015 kgCH<sub>4</sub>/kgCOD for CH<sub>4</sub> production in the sewers was calculated based on a laboratory study (Eawag 2018, Liu et al. 2015a). A study for the city of Xi'an (China) reports CH<sub>4</sub> emissions of 2.6 t for 8,705,600 inhabitants (Jin et al. 2019), which results in an EF of 0.0025 kgCH<sub>4</sub>/kgCOD, assuming a COD load of 120 gCOD/inhabitant (Gujer 2007). A comparable study for Cincinnati (USA) reported emissions of 70 kg for a catchment of 220,000 inhabitants, which results in an EF of 0.0026 kgCH<sub>4</sub>/kgCOD (Fries et al. 2018). Hence, we conclude that the value in the ER probably drastically overestimates the CH<sub>4</sub> emissions from sewers and therefore suggest to apply the average value of both studies analyzed (0.00255 kgCH<sub>4</sub>/kgCOD).

## Water line

Literature data on CH<sub>4</sub> EFs for onsite emissions from wastewater treatment (without sludge treatment) were compiled in Figure 11. The biological treatment step is the most important emission source in almost all WWTPs.

Based on the median of all involved literature data on CH<sub>4</sub> emission in the sector wastewater, an EF of 0.001 kgCH<sub>4</sub>/kgCOD is proposed (see also Figure 13). This corresponds to emissions of 1.1 kg CO<sub>2</sub>-eq per inhabitant and year when calculated with a COD load of 120 gCOD per inhabitant and day (Gujer 2007). Compared to the estimated emissions from sewer systems (~3 kg CO<sub>2</sub>-eq per inhabitant and year) the onsite emissions from WWTPs are lower.

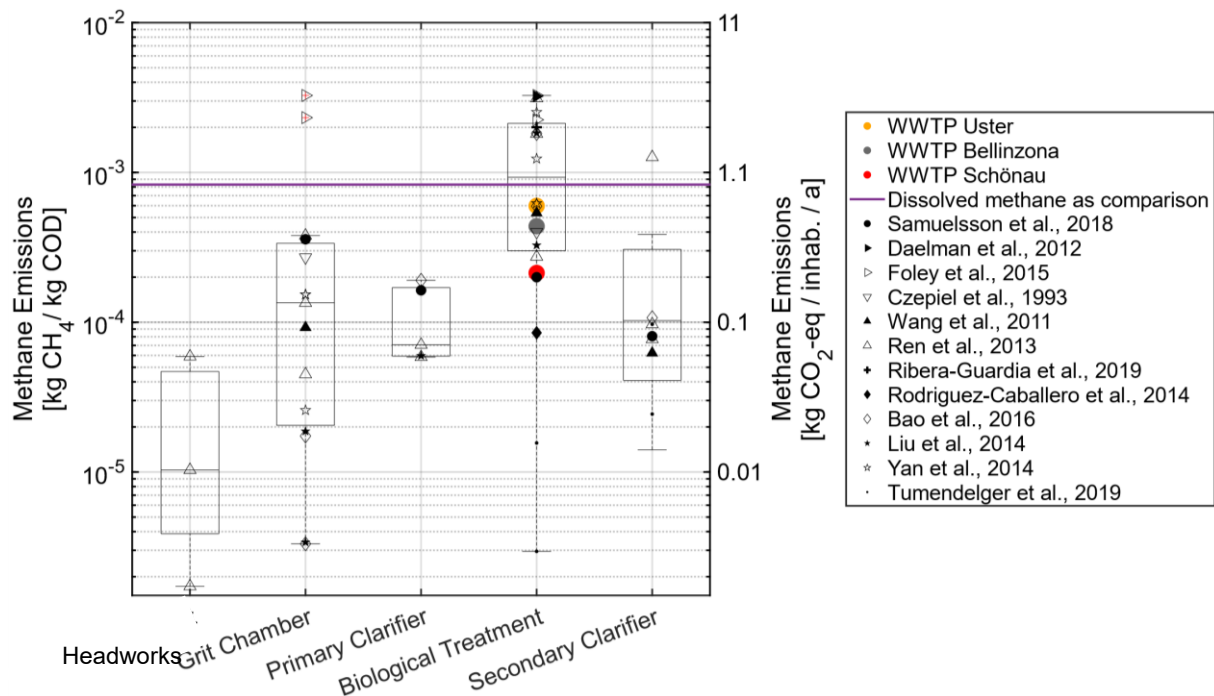


Figure 11 CH<sub>4</sub> EF for various process stages during wastewater treatment according to selected studies. Data for the Uster WWTP, Bellinzona WWTP and Schönaue WWTP were evaluated in Bühner (2020).

## Sludge line

During sewage sludge treatment, CH<sub>4</sub> formation mainly occurs in the anaerobic digestion tanks. Leakage in the sewage treatment steps (digestion, thickening, centrifugation) can lead to CH<sub>4</sub> emissions. Monitoring leakages is only possible using whole-plant measurements, which are barely able to allocate the measured emissions to a specific source within a WWTP

and therefore not further discussed here. Additionally, CH<sub>4</sub> is produced in sludge storage tanks because of the residual gas potential and the remaining organic material in the digested sludge. Very high emissions can occur in case the sludge storage tanks are open (Cunningham et al. 2015). The EFs from open sludge storage tanks were evaluated for eight WWTPs, which participated in an emission reduction project (South Pole 2014). The sludge storage tanks' emissions are generally for all WWTPs in the range between 5 % and 15 % of the total gas production (Figure 11). The mean EF of all eight investigated WWTPs is 8.3 % of the total gas production. The residual gas potential depends mainly on two parameters: the sludge retention time in the digester and the sludge retention time in the storage tank (Cunningham et al. 2015, Tauber et al. 2019).

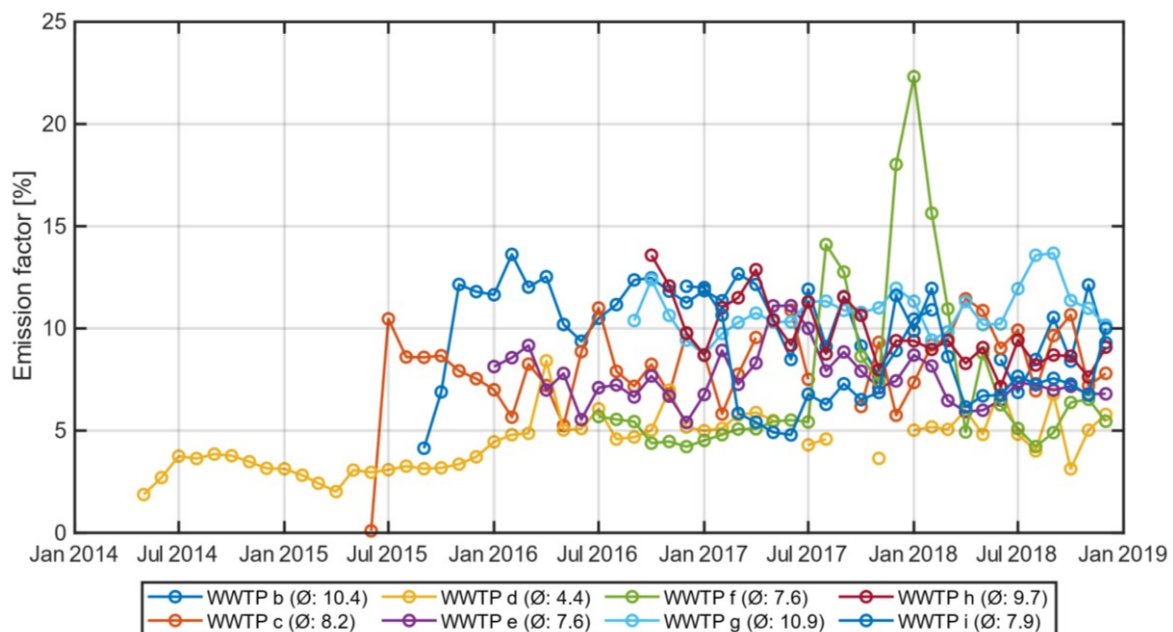


Figure 12 CH<sub>4</sub> EFs from open sludge storage tanks in 8 Swiss WWTPs given in % of the gas production according to (South Pole 2014).

The data were compared with literature data (Table 4). In general, the reported values are lower as shown in Figure 12, but still very high compared to the emissions from the water line. Tauber et al. (2019) report a much smaller EF, which could be related to the very long retention time in the digester. In summary, we conclude that open sludge storage tanks cause an important part of the total CH<sub>4</sub> emissions from sludge treatment. We expect that the share of WWTPs with open sludge tanks is decreasing as described in the ER (Eawag 2018). However,

the estimated value for 1990 (12% losses) based on an expert judgement in the ER probably overestimates the leakages.

Table 4 EFs for the residual gas potential. (HRT: hydraulic residence time; N/A: not available; FTIR: Fourier Transform Infrared)

Emission factor [%]	HRT digester [d]	HRT sludge storage tank [d]	Determination method	Reference
2 to 7	30 and 20	13 and 2.5	Simulation	Cunningham et al. (2015)
3.3	26	5	Measured in the off-gas	Daelman et al. (2012)
2.4	20	N/A	Tracer dispersion method	Samuelsson et al. (2018)
2.5	N/A	2	Batch test	Schaum et al. (2015)
0.2	42	10	Batch test	Tauber et al. (2019)
4.4 to 10.9	N/A	N/A	Measured in the off-gas	(Bührer 2020)

### ***Overall emissions (specific emission source vs. whole plant)***

To evaluate whether a bottom-up approach (sum of measured EFs from water line and sludge line) matches with whole plant measurements, we compared the available studies (Figure 13). In the bottom-up studies, emissions from sludge treatment are lower than the emission from water treatment (0.7 vs. 1.1 kg CO<sub>2</sub>-eq per inhabitant and year). This is against expectations and mainly related to sludge treatment systems monitored (Figure 13): Ren et al. (2013), Foley et al. (2015), Czepiel et al. (1993), and Wang et al. (2011) examined WWTPs without anaerobic sludge treatment. Anaerobic sewage sludge treatment are known to cause higher CH<sub>4</sub> emissions due to the substantially increased CH<sub>4</sub> production. In Switzerland, sludge treatment is mainly performed in anaerobic digestion.

Whole plant measurements resulted in significantly higher emissions and were all in a similar range. Delre et al. (2017) identified the sludge treatment as the main CH<sub>4</sub> emission source, using a tracer gas dispersion method. Bühler et al. (2021) measured total CH<sub>4</sub> emissions of two WWTPs in Switzerland with an inverse dispersion method. The results are in the same order of magnitude as the results from Daelman et al. (2012), Foley et al. (2015), Delre et al. (2017), and Samuelsson et al. (2018). Hence, we conclude that whole plant measurements

are more appropriate for a CH<sub>4</sub> EF assessment. The median value of the whole plant measurements is 0.0094 kgCH<sub>4</sub>/kgCOD.

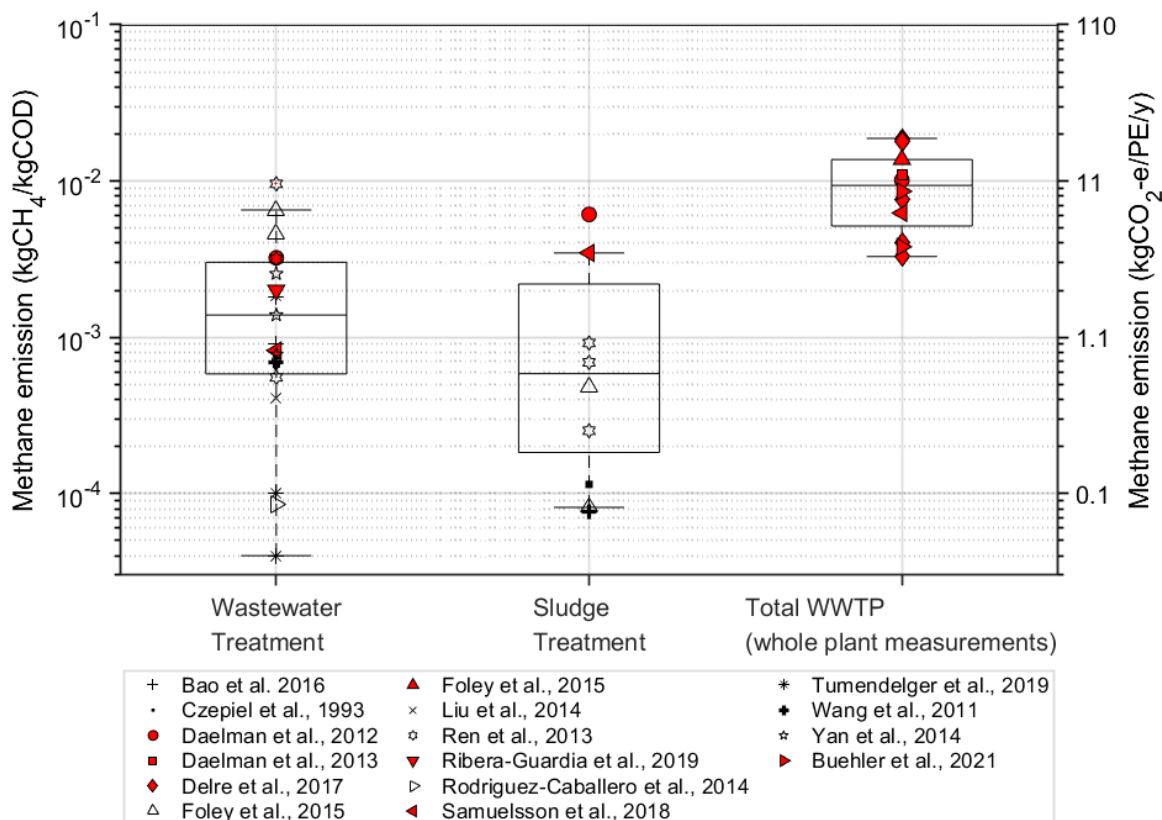


Figure 13 CH<sub>4</sub> EFs for process stages (wastewater, sewage sludge) and the total WWTP based on whole plant measurements. Red filled symbols represent WWTPs with anaerobic digestion for sludge treatment.

### 3.2.3. Proposed methodology

Given the findings from the literature review and our own measurements, we propose to apply a different method to estimate CH<sub>4</sub> emissions from WWTPs than suggested by the ER. We propose to differentiate between emissions from the sewer system, the whole WWTP and during sewage gas usage (energy line) as shown in Figure 14. Most of the emissions in the energy line (sewage gas usage) do not count to the waste sector but to the energy sector.

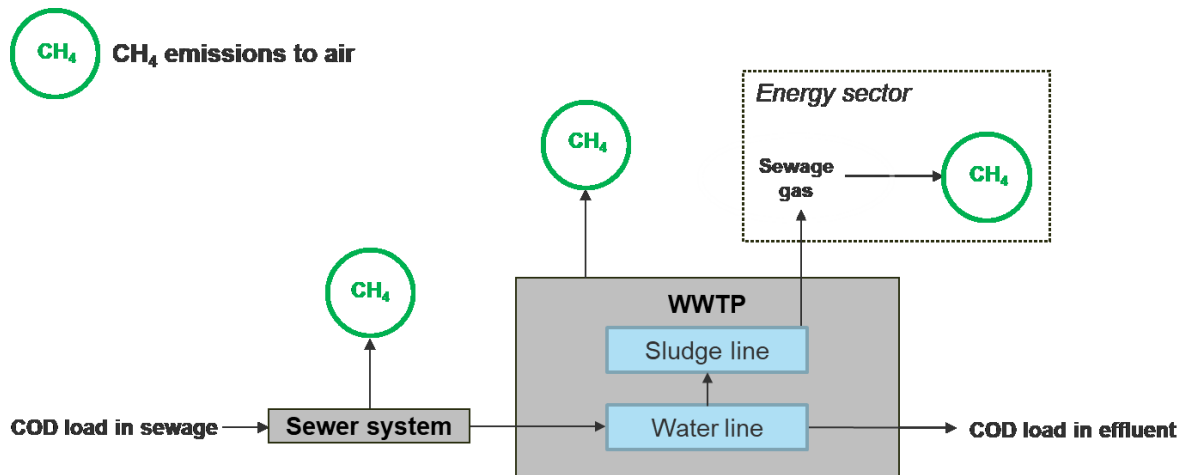


Figure 14 Proposed CH<sub>4</sub> emission model for wastewater treatment in Switzerland

### **Emissions factor sewer system ( $EF_{CH_4, SEWER}$ )**

Based on the expert review, we suggest an EF of 0.0026 kgCH<sub>4</sub>/kgCOD for the sewer system, which is the median value on the two studies described above. We do not expect any changes of the EF for the past (1990-2020) and for future estimations. We assume a high uncertainty (100%) for the EF given the relatively low data availability for representative CH<sub>4</sub> emission data from sewer systems.

### **Emissions factor wastewater treatment plant ( $EF_{CH_4, WWTP}$ )**

We suggest an EF of 0.0094 kgCH<sub>4</sub>/kgCOD for the whole wastewater treatment (water line + sludge line) based on the literature review. Although a reduction of emissions can be achieved by the coverage of sludge storage tanks, we suggest to omit the reduction of the EF over time as proposed by the ER, since the representativity of the selection of whole-plant measurements for Switzerland cannot be quantified for a specific year due to missing data on sludge treatment in Switzerland. Assuming a reduction factor for the average EF assessed would be therefore arbitrary and most accurate emissions factors are reached by using the median of the studies analyzed. The uncertainty of the EF is calculated with the standard mean error (0.0015 kgCH<sub>4</sub>/kgCOD) of the available whole-plant studies. By multiplying with the coverage factor  $k$  (2.2 for 11 degrees of freedom), the uncertainty equals to 0.0034 kgCH<sub>4</sub>/kgCOD or 36% of the median value.

### ***Emissions factor energy line***

The EFs of sewage gas usage are estimated as in previous versions of the NIR (FOEN 2020a) based on the federal statistics (SFOE 2017). Only the emissions caused by leakage, sewage gas upgrade and combustion in torches are accounted to the waste sector. Uncertainties are not considered for these variables.



## 4. Emission estimation

### 4.1. N<sub>2</sub>O emissions

#### 4.1.1. Proposed methodology

Total N<sub>2</sub>O emissions from WWTPs are calculated using input data and equations defined in chapter 2.1.3 and 3.1.3. The resulting equations are used to calculate the total emissions from WWTPs (Equation 6 and 7).

Nitrous oxide emissions from WWTPs to the atmosphere:

$$N_2O_{PLANT} = N_{INFLUENT} * EF_{N_2O, PLANT} \quad (6)$$

Nitrous oxide emissions from the effluent :

$$N_2O_{EFFLUENT-WATERBODY} = N_{EFFLUENT} * EF_{N_2O, EFFLUENT-WATERBODY} \quad (7)$$

Output variables:

$N_2O_{PLANT}$  Nitrous oxide emissions from WWTPs [kg N<sub>2</sub>O-N / year]

$N_2O_{EFFLUENT-WATERBODY}$  Nitrous oxide emissions from effluent [kg N<sub>2</sub>O-N / year]

Input variables:

$EF_{N_2O; PLANT}$  Emission factor for emissions of the plant [kg N<sub>2</sub>O-N / kg N]

$EF_{N_2O; EFFLUENT-WATERBODY}$  Emission factor for emissions of the effluent [kg N<sub>2</sub>O-N / kg N]

#### 4.1.2. Comparison with IPCC methodology

To demonstrate the value of a country-specific methodology for the estimation of N<sub>2</sub>O emissions from WWTP, we calculated the total N<sub>2</sub>O emissions from WWTPs using the 2006 and the 2019 IPCC methodologies as well as our proposed methodology (see 4.1.1). The resulting emissions for the year 2019 (Figure 15) show that the 2006 IPCC methodology drastically underestimates the total emissions and unrepresentatively estimates the origin of the emissions (onsite vs. from effluent). The differences according to the 2019 refinements are somewhat smaller and mostly due to the lower EF for onsite emissions (1.6% vs 2.5% in IPCC 2019). The full data series are available in the Appendix ('Calculations CH' file). The calculated

uncertainties of the total N<sub>2</sub>O emissions calculated with our method are 40% (see Appendix, 'Additional data' file).

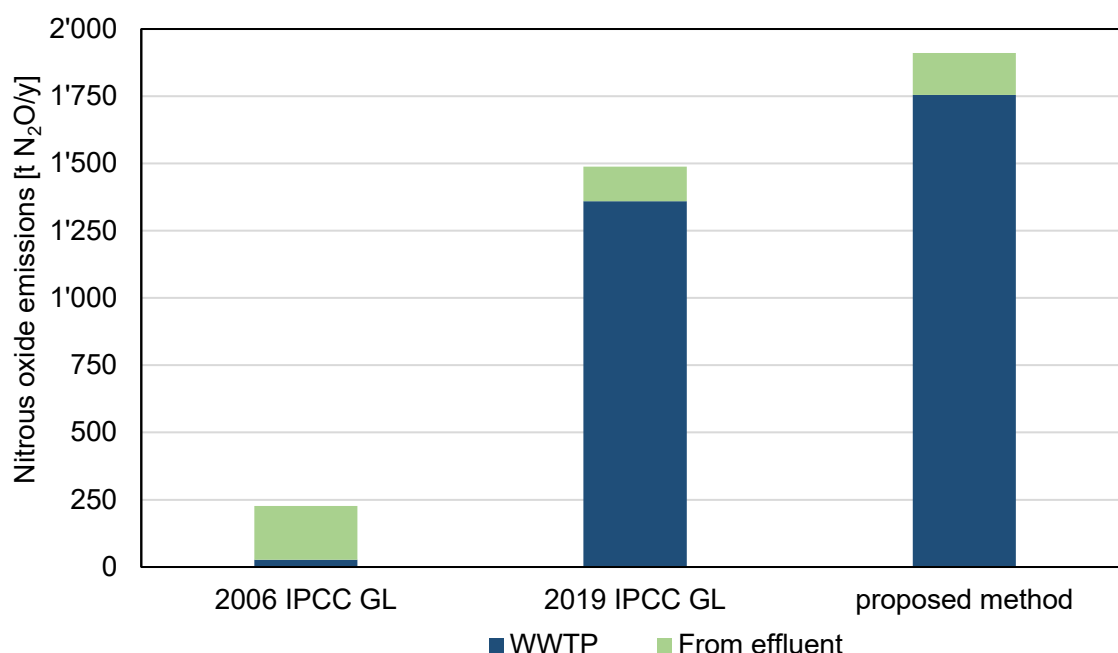


Figure 15 Estimated Swiss N<sub>2</sub>O emissions from source category 5D calculated with different methods for 2019

According to our estimations, the overall emissions of N<sub>2</sub>O generally decreased since 1990 because of the increasing nitrogen removal rates at WWTPs (Figure 16). The decrease can be linked to two effects. Firstly, WWTPs with full nitrogen removal were shown to have lower N<sub>2</sub>O EFs (Table 3). Secondly, increased nitrogen removal reduced nitrogen discharge to natural waterbodies and subsequent emissions of N<sub>2</sub>O. The share of nitrogen removing WWTPs will further increase given recent federal regulations (Swiss Parliament 2021). As a consequence, N<sub>2</sub>O emission from WWTPs are expected to further drop after the stagnation period of the overall nitrogen removal rate between 2010 and 2025 (Figure 5).

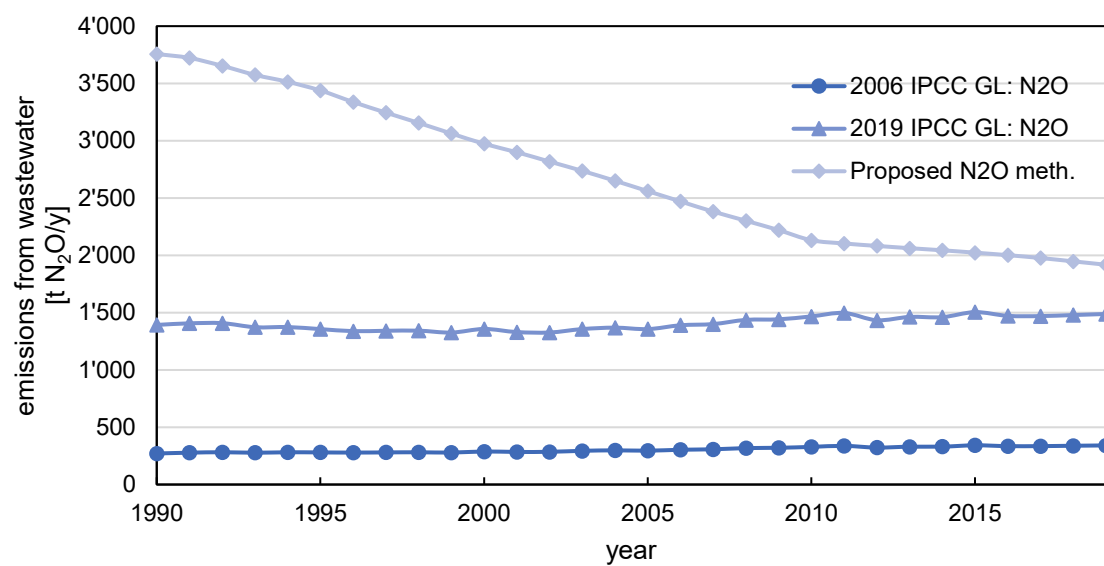


Figure 16 Estimated Swiss N<sub>2</sub>O emissions from source category 5D according to different methods for 1990–2019

## 4.2. CH<sub>4</sub> emissions

### 4.2.1. Proposed methodology

Total CH<sub>4</sub> emissions from WWTPs are calculated using input data and equations defined in chapter 2.2.2 and 3.2.3. The resulting equations are used to calculate the total emissions from WWTPs (Equation 8, 9 and 10).

CH<sub>4</sub> emissions from sewer system:

$$CH_{4, SEWER} = TOW_{SEWER} * EF_{CH_4, SEWER} \quad (8)$$

CH<sub>4</sub> emissions from wastewater treatment (water line + sludge line):

$$CH_{4, WWTP} = TOW_{SEWER} * EF_{CH_4, WWTP} \quad (9)$$

CH<sub>4</sub> emissions from sewage gas usage:

$$CH_{4, SEWAGE GAS} = \sum P_{j, SEWAGE GAS} * EF_{CH_4, SEWAGE GAS, j} \quad (10)$$

Output variables:

CH <sub>4, SEWER</sub>	CH <sub>4</sub> emissions from sewer system [kg CH <sub>4</sub> / year]
CH <sub>4, WWTP</sub>	CH <sub>4</sub> emissions from WWTPs [kg CH <sub>4</sub> / year]
CH <sub>4, SEWAGE GAS</sub>	CH <sub>4</sub> emissions from sewage gas usages / leakages [kg CH <sub>4</sub> / year]

Input variables:

TOW <sub>SEWER</sub>	Organically degradable material in sewer [kg COD / year]
EF <sub>CH<sub>4</sub>, SEWER</sub>	Emission factor for CH <sub>4</sub> emission from the sewer system [kg CH <sub>4</sub> / kg COD]
EF <sub>CH<sub>4</sub>, WWTP</sub>	Emission factor for CH <sub>4</sub> emission from wastewater treatment [kg CH <sub>4</sub> / kg COD]
P <sub>j, SEWAGE GAS</sub>	Sewage gas usage [TJ / year]
EF <sub>CH<sub>4</sub>, SEWAGE GAS</sub>	Emission factor for CH <sub>4</sub> from sewage gas usage [kg CH <sub>4</sub> / TJ]

#### 4.2.2. Comparison with IPCC methodology

To demonstrate the value of a country-specific methodology for the estimation of CH<sub>4</sub> emissions from WWTP, we calculated the total CH<sub>4</sub> emissions from WWTPs using the 2006 IPCC methodology as well as our proposed methodology (see 4.2.1). We do not include the 2019 IPCC refinements in the comparison, since there are no major changes compared to the 2006 IPCC methodology. The resulting emissions for the reference year 2019 (Figure 17) show that the 2006 IPCC methodologies overestimate the total emissions and wrongly project the origin of emissions (Figure 18). The full data series are available in the Appendix ('Calculations CH' file).

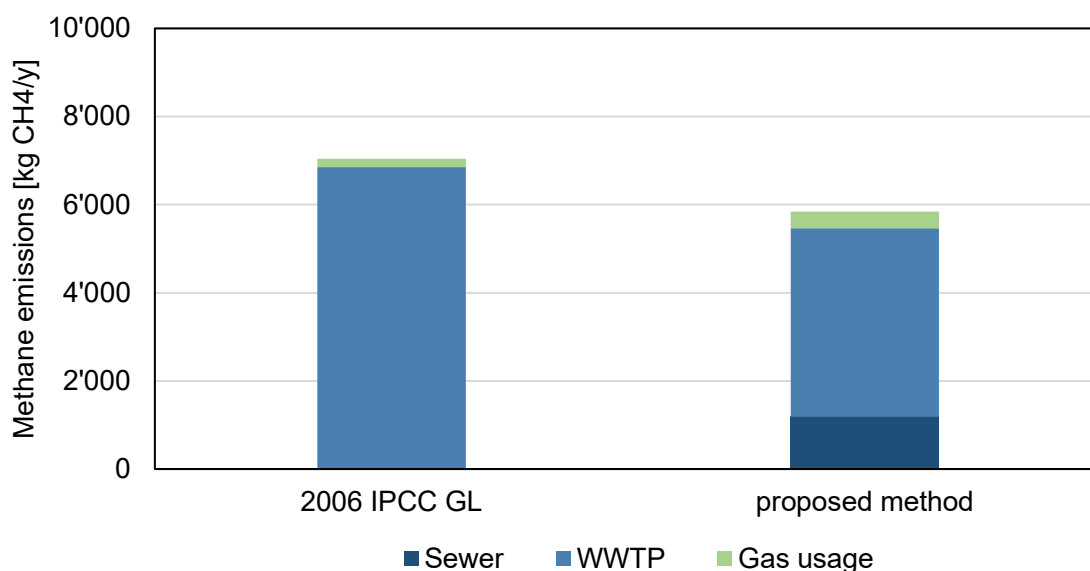


Figure 17 Swiss CH<sub>4</sub> emissions from source category 5D calculated with different methods for the year 2019

According to our estimations, the overall emissions of CH<sub>4</sub> from wastewater treatment increased in accordance with the population growth (Figure 17). The overall uncertainties are relatively high (103%, see appendix 'Additional data' file) because data availability is relatively weak for sewer systems.

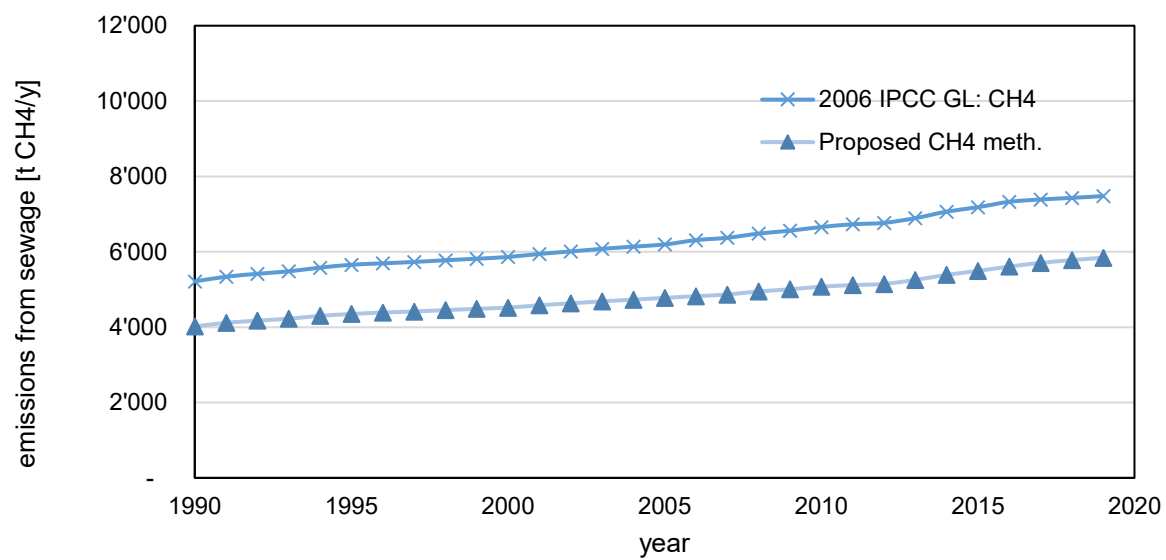


Figure 18 Estimated Swiss CH<sub>4</sub> emissions from source category 5D according to different methods for 1990 to 2019.

## **5. Suggestions for further improvements**

### **5.1. N<sub>2</sub>O emissions**

Estimations of N<sub>2</sub>O emissions from WWTPs could be further improved using nitrite effluent data, since nitrite was shown to be a good proxy for N<sub>2</sub>O emissions (Gruber et al. 2020). However, gathering enough data is challenging, since nitrite is not always monitored on Swiss WWTPs and data is not collected by FOEN (e.g. no entries in FOEN (2021a)).

The ratio of emissions from unaerated process stages in WWTPs could be improved by performing monitoring campaigns. Similarly, the EF for N<sub>2</sub>O from natural water bodies could be improved by performing monitoring campaigns in Switzerland similar to the study by Marescaux et al. (2018). Both approaches are linked to substantial costs.

### **5.2. CH<sub>4</sub> emissions**

The highest uncertainties in the CH<sub>4</sub> emission estimations are linked to the EFs from sewer systems and the CH<sub>4</sub> losses during sludge storage. To improve EF from sewers further monitoring campaigns of CH<sub>4</sub> emissions from sewers applicable to Switzerland are required. For an improved estimation of CH<sub>4</sub> losses from WWTPs, additional whole-plant CH<sub>4</sub> emission measurements are needed to cover all potential individual emission sources: the water line, the sludge line and the energy line including leakages and slurry storage. In addition, separate measurements of emissions from sludge storage based on Integrated Horizontal Flux (IHF) or Mass Balance Method are recommended according to (Kupper et al. 2018)

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