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Greenhouse Gas Emissions from Wastewater Treatment Plants

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Abstract

Direct and indirect greenhouse gases emissions of two model municipal WWTPs were estimated using carbon footprint analyses. One WWTP was designed with anaerobic digestion, the other one with simultaneous aerobic stabilization of sewage sludge. Emission factors were derived from literature values. For direct nitrous oxide emissions, a new estimation model based on measurements at eight Austrian WWTPs was applied. Results show that the direct nitrous oxide emission from the activated sludge tanks dominates the carbon footprint of WWTPs with a moderate nitrogen removal. Anaerobic digesters and anaerobic sludge storage tanks can also become a relevant source of direct methane emissions.

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Keywords: Greenhouse gases; carbon footprint; wastewater treatment; nitrous oxide; methane.

1. Introduction

Municipal wastewater collection and disposal at wastewater treatment plants (WWTP) contribute to the emission of greenhouse gases (GHG) in the atmosphere. This is also highlighted in the Intergovernmental Panel on Climate Change (IPCC) guidelines, used for national GHG inventories within the member countries [1]. Carbon footprint analyses have been shown to be applicable to estimate GHG emissions also in the wastewater sanitation sector [2,3].

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The carbon footprint represents the total set of GHG emissions caused directly or indirectly by an activity or resulting from the different life cycle stages of a product. The global warming potential (GWP) of GHGs is referred to carbon dioxide (CO_2) as reference gas and usually expressed as equivalent carbon dioxide (CO_2e).

Emission paths in wastewater sanitation are exemplary summarized in Table 1. Direct GHG emissions include emissions of methane (CH_4) and nitrous oxide (N_2O) that can be biologically produced and emitted in sewers and at WWTPs during wastewater and sewage sludge treatment. CO_2 is also emitted at WWTPs but it is usually not considered in the CO_2-balance, being predominantly biogenic and therefore climate neutral. Indeed, isotopic mass balance analyses revealed that 4 to 14% of the total organic carbon in wastewater influent can be of fossil origin [4]. Indirect GHG emissions occur at WWTPs mainly by the consumption of electricity, by burning fossil fuel for transportation, by the use of chemicals (e.g. for phosphate precipitation and sludge dewatering) and by the disposal of sewage sludge (biosolids).

The informative value of carbon footprint analyses relies on a well-considered selection of the emission factors (EF) used for the calculations. Several sources and databases are currently available, especially for indirect emissions [5,6,7]. For direct emissions of CH_4 and N_2O at WWTPs literature values can be applied [8,9,10]. Few field measurements have been conducted so far on CH_4 emissions. In contrast, direct N_2O emission from activated sludge tanks have been intensively investigated in the last decade. Despite of this, the estimation of representative EFs for N_2O emission is still hampered by the wide variability range of the measurement results. This can be attributed to the strong influence of operating conditions (loading rate, dissolved oxygen concentration, temperature, etc.) on N_2O production and emission. A new estimation model based on long-term measurements at eight Austrian WWTPs can help improving the estimation of direct N_2O emissions from activated sludge tanks [11]. Innovative in the model is the definition of EFs as a function of the operating condition at every single WWTP. In particular, the degree of TN removal showed a good correlation with direct N_2O emissions. This new approach for estimating direct N_2O emission was applied in the present study to calculate the carbon footprint of two model municipal WWTPs. Additionally, the impact of process configuration - with and without anaerobic digestion of sewage sludge - on the CO_2 balance of WWTPs was evaluated.

Table 1. Direct and indirect GHG emissions from wastewater collection, treatment and discharge [2].

Direct GHG emissions	Indirect GHG emissions
Wastewater collection (sewer system)	Electricity supply
Wastewater treatment (WWTP)	Transportation (e.g. chemicals, sewage sludge)
Wastewater discharge in water bodies	Use of chemicals and additives (including GHG emissions in the upstream stages of production)
	Disposal/reuse of residuals (e.g. biosolids)

2. Methods

The carbon footprint analyses were performed for two model municipal WWTP of 50,000 population equivalents (PE) each. The balance boundary was set by the sewage sludge treatment, still including anaerobic digestion and sludge dewatering. In contrast, the following processes were not considered in the CO_2-balance:

- GHG emissions resulting from the disposal of biosolids: this specific topic is discussed elsewhere [12, 13].
- GHG emissions from the sewer system: the range of CH_4 and N_2O emissions in sewer systems is influenced by several factors (e.g. sewer type, lengths, slope, sewage temperature) and still controversial. In contrast to the assumption of the IPCC guidelines postulating that CH_4 und N_2O emissions in open sewers are negligible [1], field measurements revealed that emissions can be significant [14, 15].
- GHG emissions during the construction phase of WWTPs: in this regards [16] showed that GHG emissions resulting from the use of raw materials and energy in the construction phase of two real WWTPs correspond to 10-20% of the total GHG emission of the WWTP for a service life of 30 years.

2.1. Model WWTPs

Both model WWTPs were designed as single stage activated sludge plants, one with anaerobic digestion of sewage sludge (WWTP-AD), the other one with simultaneous aerobic sludge stabilization (WWTP-SAS). The influent wastewater flow rate and pollutant load was calculated using pro capita loading values typical for municipal wastewater in Central Europe: 120 g COD/PE/d; 11 g TN/PE/d; 1,6 g TP/PE/d; 200 L/PE/d [17].

Relevant specifications of both WWTPs are summarized in Table 2. Assumption were based on the results of a broad benchmarking study comprising 96 Austrian WWTPs [18] and literature value [32]. Simultaneous aerobic sludge stabilization is characterized by an intrinsic higher energy demand for aeration (+9 kWh_{el}/PE/a) and specific biosolids production (+5 g Total Solids/PE/d). Nitrogen (TN) is removed from wastewater by nitrification and denitrification. It was assumed that TN removal at WWTP-AD exhibits an unexploited optimization potential and that it can be increased from 77% to 90% by setting targeted operating measures. Due to the lower COD availability for denitrification a somewhat lower degree of TN removal can be achieved at WWTP-AD, even after optimization.

Postulating an electric efficiency of the block heat and power unit of 30%, 15 kWh_{el}/PE/a electricity can be produced at WWTP-AD out of a daily biogas production of 23 L per PE (CH_4-content of biogas: 60%). The provided electricity can cover the energy demand for aeration in the activated sludge tank.

Table 2. Specifications of the model wastewater treatment plants

	WWTP-AD	WWTP-SAS	Unit
COD influent WWTP	120	120	g COD/PE/d
COD primary sludge	42	-	g COD/PE/d
COD excess sludge	29	38	g COD/PE/d
COD effluent WWTP	5	4	g COD/PE/d
COD oxidized	44	78	g COD/PE/d
TN nitrified	9.3	8.5	g N/PE/d
TN denitrified	7.1*	7.9	g N/PE/d
COD removal	96	97	%
TN removal	77 (90)**	92	%
TP removal	90	90	%
Energy demand for aeration	15	24	kWh_{el}/PE/a
Energy demand for WWTP	30	37	kWh_{el}/PE/a
COD in methane	40	-	g COD/PE/d
Methane production	14	-	L***/PE/d
Biogas production	23	-	L/PE/d
Electricity from biogas	15	-	kWh_{el}/PE/a
Sludge production	37	42	g TS/PE/d

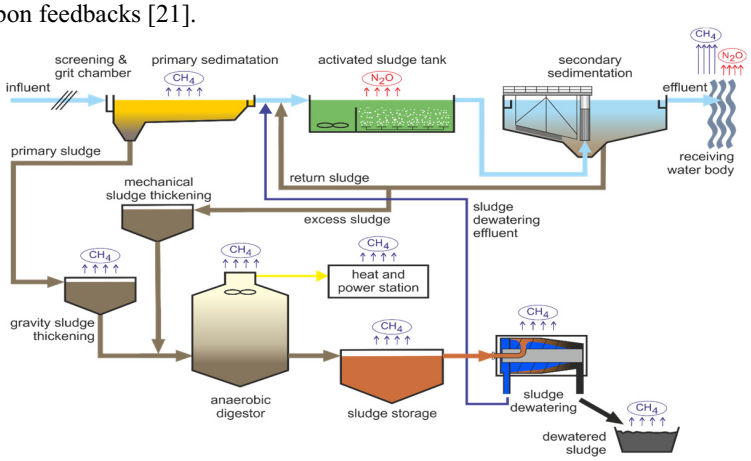
*By 77% TN removal; **TN removal can be increased to 90% optimizing the operation; ***gas standard volume: 273.15 °K, 1.01325 bar

2.2. Direct GHG emissions from WWTPs

The production and emission of CH_4 and N_2O at WWTPs can occur at different treatment stages (Fig. 1). N_2O is produced mainly in the activated sludge tank as a side product of the biological TN removal by nitrification and denitrification and then stripped into the atmosphere by aeration [8, 10]. At WWTPs with anaerobic digestion primary sedimentation and the whole sludge line can be a potential source of direct CH_4 emissions [19]. Aerobically stabilized sewage sludge can also become a source of CH_4 as soon as anaerobic conditions are established, e.g. during long term storage for several months [20]. The COD and TN leaving the WWTP with the treated effluent can promote further CH_4 and N_2O production and emission in the receiving water bodies, depending on existing milieu

conditions. The calculation of the climate impact of N_2O and CH_4 was performed applying the GWP of 25 kg CO_2e /kg CH_4 and of 298 kg CO_2e /kg N_2O respectively, referring to a time framework of 100 years and including climate carbon feedbacks [21].

a)



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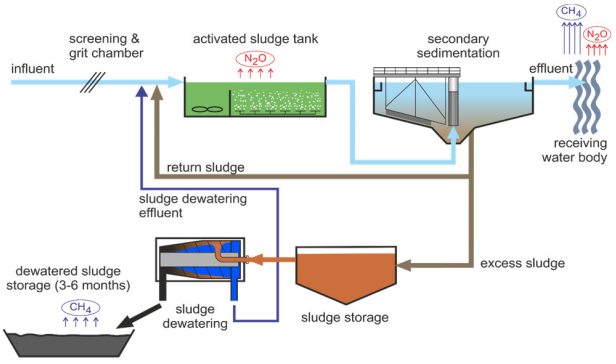

Fig. 1. (a) GHG emissions at WWTP-AD with anaerobic digestion and (b) at WWWTP-SAS with simultaneous aerobic sludge stabilization.

2.3. Estimation of N_2O emissions from municipal WWTPs

Activated sludge tanks have shown to be the main sources of N_2O at WWTP. A more detailed discussion on the formation mechanisms of N_2O emissions in activated sludge tank can be found in literature [8, 10]. According to current understanding, N_2O production and emission during nitrification can be reduced by optimizing process conditions but not completely avoided. This applies to both postulated formation paths, the nitrifier denitrification and the oxidation (biological or chemical) of hydroxylamine. During denitrification N_2O is produced as obligate intermediate. However, under favorable process conditions (e.g. low dissolved oxygen and nitrite concentrations, sufficient COD availability) denitrification can become a significant N_2O sink promoting the reduction of N_2O to gaseous N_2.

The variability range of direct N_2O emissions measured at WWTPs so far is wide. EFs expressed as kg N_2O-N emitted per influent kg TN vary between 0.003 und 2.6 % [11]. This pronounced variability mainly derives from the significant impact that operating conditions have on N_2O production [8,22]. The degree of TN removal and the loading conditions in activated sludge tanks were identified as the major operating parameters affecting direct N_2O emission within long term measurement campaigns at eight municipal WWTPs in Austria [11, 23]. The measurements took place in activated sludge tanks applying long-term online measurements in the exhausted aeration air (floating gas hood connected to a IR-Spectrometer) as well as in the bulk liquid (Unisense micro-sensor) over several weeks. A decreasing EF (g N_2O-N/ g $N_{influent\ WWTP}$) was coupled with an increasing TN removal

efficiency and a decreasing volumetric loading rate (Fig. 2). The observed correlation with the TN removal performance confirms the role of the denitrification as N_2O sink in activated sludge tanks. Considering that lower TN removal were mostly coupled with high volumetric loading rates in the activated sludge tanks, the regression model reflects also the influence of this operating parameter. Based on the project results an Austrian country-specific estimation method of direct N_2O emissions from activated sludge tanks depending on TN removal was developed [11, 24], in objection to the less representative EF of 3.2 g N_2O/person/a of the IPCC guidelines [1]. The IPCC EF is based on measurements (grab samples, no online measurements) performed at only one single WWTP of not specified TN removal efficiency and not receiving any wastewater from industrial sources [25].

The direct N_2O emissions from the activated sludge tanks of both model WWTPs were calculated using the regression model depicted in Fig. 2. At WWTP-AD, achieving a TN removal of 77%, 0.75% of $TN_{influent}$ is emitted in the atmosphere as N_2O. At WWTP-SAS the EF decreases to 0.05% due to the much higher TN removal of 92%. For N_2O emissions occurring in receiving water bodies the IPCC EF of 0.5% of effluent TN was applied [1].

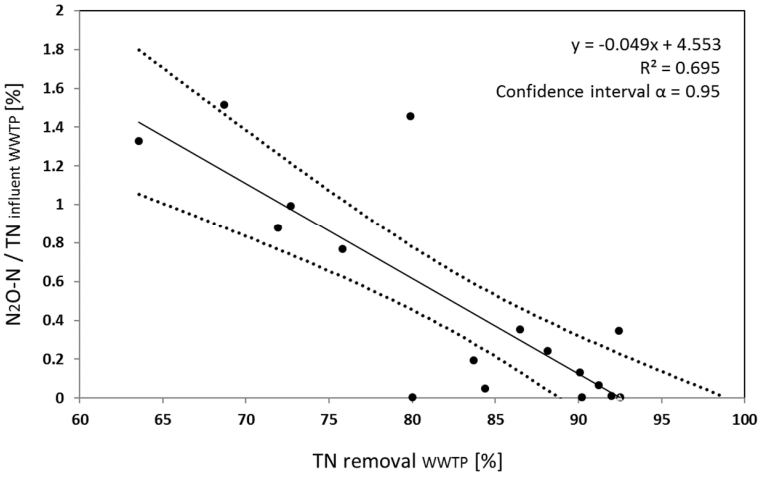

Fig. 2. Scatter plot of direct N_2O emissions from activated sludge tanks (g N_2O-N/g $N_{influent\ WWTP}$*100) and TN removal efficiency (%) comprising 18 measurement campaigns at eight Austrian WWTPs [11]. Each point in the diagram represents the average overall direct N_2O emissions of activated sludge treatment resulting from every single measurement campaign.

2.4. Estimation of CH_4 emissions from municipal WWTPs

The state of knowledge of direct CH_4 emissions at WWTPs is not so comprehensive as for N_2O, since few field measurements have been published in literature so far [9, 19, 26]. At WWTPs with anaerobic digestion about 75% of the direct CH_4 emissions are expected to occur in the sludge line [19]. Potential sources are anaerobic digesters (fugitive biogas emissions), sludge storage tanks and the slip of the block heat and power plant. In the water line CH_4 can be formed in the primary sedimentation and then stripped out with the aeration air in activated sludge tanks. Under aerobic conditions CH_4 can be also partially biologically oxidized, as observed by [19].

CH_4 emission factors for anaerobic digesters from literature range between 0% [27] and 11% [28] of the produced CH_4, the highest number being merely based on expert judgement. Depending on the sludge stabilization degree achieved in the digester, CH_4 emissions by storing and dewatering of digested sewage sludge range between 2% and 4.5% of the CH_4 production [19, 29]. In comparison to that, the release of CH_4 dissolved in the digested sludge is of less relevance (<<1%), as confirmed by measurements in digested sludge of seven different municipal WWTPs [30]. The percentage of unburned CH_4 measured in the combustion air of block heat and power plant vary between 1.3 % [19] and 1.8 % [31] of the CH_4 produced.

Measurements at WWTPs using an integrated remote sensing approach (e.g. dynamic tracer dispersion method) provided CH_4 EFs for the whole WWTP up to 30% of the biogas production [26]. For the model WWTP-AD a more conservative total CH_4 emission of 10.4% was assumed. Referred to the influent COD load this amounts to

8.7 g CH_4/kg $COD_{influentWWTP}$, which is in the range of the value reported by [9].

According to the IPCC Guidelines [1] CH_4 emissions at WWTPs with simultaneous aerobic sludge stabilization are negligible as long as the activated sludge tanks are not overloaded and sufficiently aerated. This assumption applies for the model WWTP-SAS. The CH_4 emissions resulting from the long-term storage of dewatered sludge (> 3-6 months) were calculated on the basis of a further COD degradation under anaerobic conditions of 6%. For CH_4 emissions in the receiving water bodies, the IPCC EF of 0.025 kg CH_4/kg $COD_{effluent}$ was adopted.

2.5. Estimation of indirect GHG emissions of municipal WWTPs

Indirect GHG emissions were estimated using EFs available in reliable data bases [5,6,7] as depicted in Table 3. The whole upstream chain of chemical application at WWTPs (extraction of raw material, production and distribution) was considered. It can be noted that the manufacturing process can significantly impact the EF of chemicals. The production of $FeCl_3$ out of iron oxide and hydrochloric acid is e.g. more emission intensive than using pickling acids and chlorine gas (Table 3). In case a product is not specifically produced but results as a side product out of an industrial manufacturing process, GHG emissions need to be reallocated as suggested by [33].

At the model WWTPs phosphate precipitation is achieved by adding iron chloride in excess (1.5 mol Fe^{3+} per mol PO_4^{2-}). Sewage sludge dewatering occurs in a filter press chamber by adding polymers (7 kg active substance per ton total solids) and iron chloride (60 kg $FeCl_3$ per ton TS). The EF for iron chloride was assumed to be 0.05 kg CO_2e/mol Fe^{3+} [7], for polymeres 2.62 kg CO_2e/kg active substance [7]. CO_2e emissions resulting from chemicals transportation were also included in the carbon balance.

Table 3. Emission factors (as CO_2e) used for the calculation of indirect GHG emissions at the model WWTPs.

	Emission factor	Unit	Source
Electric energy generation and supply in Austria	0.38	kg CO_2e/kWh	[5]
Transport by lorry 16-32 t, EURO5	0.17	kg CO_2e/(ton km)	[5]
Iron chloride for phosphate precipitation:			
• Iron oxide and hydrochloric acid	0.160	kg CO_2e/mol Fe^{3+}	[7]
• Pickling acids and chlorine gas	0.055	kg CO_2e/mol Fe^{3+}	[7]
Sludge conditioning (thickening, dewatering)			
• Organic polymers	2.62	kg CO_2e/kg $_{active\ substance}$	[7]
• Organic polymers	1.18	kg CO_2e/kg $_{active\ substance}$	[6]

3. Results

The carbon footprint analysis for WWTP-AD results in a total emission of approx. 36 kg CO_2e/PE/a (Fig. 3). The CO_2e credit of 5.9 kg CO_2e/PE/a provided by the energetic use of the biogas is already considered. This reduction potential of CO_2e emissions is offset by 9.5 kg CO_2e/PE/a emitted directly at the plant as CH_4, according to the assumption made in chapter 2.4. The impact of indirect CO_2e emission caused by the electricity consumption of the WWTP is much lower compared to the direct N_2O-Emission (16 vs. 43% of the total emission). In case the TN removal at the WWTP is increased from 77% to 90% by optimizing the operation of the activated sludge process, direct N_2O emission could be significantly reduced from 15.5 to 4.3 kg CO_2e/PE/a, as predicted by the regression model in Fig. 2. This would lead to an additional reduction of the electricity demand for aeration (0.3 kg CO_2e/PE/a) as well as to a further decrease of N_2O emitted in the receiving water bodies, through the enhanced effluent quality of the plant (1.2 kg CO_2e/PE/a). The carbon footprint of WWTP-AD could be improved from 36 to approx. 24 kg CO_2e/PE/a.

The calculated carbon footprint of WWTP-SAS is slightly lower, amounting to approx. 24 kg CO_2e/PE/a. Therein, 60% of the CO_2e emission is caused by electricity consumption. Due to the higher degree of TN removal (92%) achieved at this plant a much lower N_2O emission can be expected (1.3 kg CO_2e/PE/a). CH_4 emissions arising from the long term storage of dewatered biosolids contribute with 4.6 kg CO_2e/PE/a (19%) to the total emission.

As a result of the high treatment capacity achieved at both WWTPs (low COD and TN effluent load) N_2O and

CH_4 emissions in the receiving water bodies play a minor role in the CO_2e balance (9% of total CO_2e emission at WWTP-AD and 7% at WWTP-SAS). CO_2e emissions from indirect sources (chemicals and transportation) contribute as well to a less degree to the carbon footprint.

The comparison between the two model WWTPs clearly show that – under the assumptions made – the carbon footprint of WWTPs with anaerobic digestion resembles the one of WWPTs with simultaneous aerobic sludge stabilization, provided that the degree of TN removal is comparable and CH_4 losses from the sludge stabilization can be reduced. Through an improved TN removal several advantages can be gained: direct N_2O emissions can be reduced, the energy demand for aeration can be decreased and a higher effluent quality can be achieved. The minimization of direct CH_4 emissions not only improves the carbon footprint of WWTPs but also increases the energy yield by combusting this renewable energy carrier in a block heat and power unit.

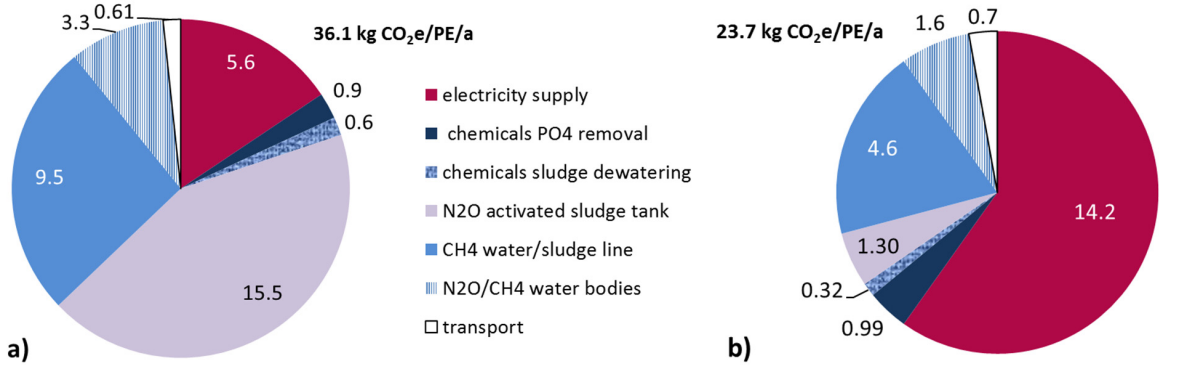

Fig. 3. Results of the carbon footprint analyses for the WWTP (a) with anaerobic digestion and (b) with simultaneous aerobic sludge stabilization.

4. Conclusions

The results of the carbon footprint analyses show that the direct N_2O and CH_4 emissions significantly impact the CO_2e balance of municipal WWTPs. This impact will become even more pronounced when in the future electricity provided by the grid will be generated using exclusively renewable sources.

The choice of EFs strongly impacts CO_2e balances and therefore requires a careful professional approach. In this regard, sensitivity analyses can help to assess the confidence level of the CO_2e balance. Despite the strong variability of the measured EF for direct N_2O emissions at WWTPs, the knowledge of the production mechanisms and influencing factors has been matured so far, that estimations based on the influent nitrogen load and on the nitrogen removal of the WWTP are now possible. In contrast, the knowledge of the reduction potential of direct CH_4 emission at WWTPs is still inadequate and requires further research. Current results indicate that a careful plant design and an optimized operation of activated sludge treatment in terms of TN removal and use of chemicals are prerequisites for a low carbon footprint of WWTPs. However, it has to be emphasized, that carbon footprint analyses alone cannot provide the basis for decision on wastewater sanitation matters. An ecological evaluation in the context of a comprehensive life cycle assessment (LCA) represents a valuable addition.

The carbon footprint analyses reveal that GHG emissions from municipal WWTPs have a small impact at global scale, corresponding in average to 0.8% of the total CO_2e emission in Austria in 2013 [24] and 0.45% of the yearly average pro capita CO_2e emission in Europe [34], respectively. Nevertheless, the optimization of WWTP operation can be significant at local scale and help improving the carbon footprint of urban areas.

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