Exhaust Emissions of Biogas-Driven Combined Heat- and Power Plants

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In order to determine the actual volume of emissions from agricultural biogas-driven heat- and power plants, the exhaust emissions of selected practically operated plants with pilot-injection engines were measured over a longer period. These measurements showed that none of the units measured entirely met the emission limits of the TA Luft (Technical Regulations Concerning Air Pollution). At the discretion of the licensing authorities, these limits for plants with a thermal firing output of a total of 1 MW or more can also be applied in construction permit procedures for smaller plants like those frequently used in practice for agricultural biogas combustion. If necessary, however, more practice-oriented emission limits like those recommended by the Bavarian Environmental Protection Agency must be employed in such cases. Optimized engine settings and regular maintenance could significantly reduce the emission potential.

Keywords
Exhaust fumes, emissions, biogas, combined heat- and power plant

Introduction

With the foreseeable depletion of fossil energy resources and the greenhouse problems, the use of regenerative energies is increasingly gaining in importance. In recent years, stronger efforts have been made to find alternatives to fossil energy carriers. The utilization of biogas in combined heat- and power plants is one of these alternatives. The advantage of this technology resides in the possibility of power-heat combination, where the waste heat of the engine as a by-product of electricity generation is utilized. Hence, the total efficiency of the process is increased. As a result of the introduction of the Electric Power Input Act in 1991 and the Renewable Energy Act in the year 2000, the combined heat- and power plant technology has significantly gained in acceptance. Combined heat- and power plants can be operated in a wide output range between a few kW and several hundred kW [1]. This possibility of output adaption to the individual requirements provides the basis for the extension of decentralized energy supply. In Bavaria, a considerable increase in the number of newly built and extended plants was observed [2]. Given these considerations, the exhaust gases produced during biogas combustion in the engines is increasingly gaining in importance. In the agricultural area, mainly two engine concepts are employed: the petrol engine, which works according to the spark-ignition gas principle and is run with pure gas and diesel engines based on the pilot-injection principle and operated with a fuel oil additive (igniter oil). During the combustion process in the engine, noxious exhaust gases are produced in addition to heat and energy. In the case of biogas engines, carbon monoxide (CO), nitric oxides (NO_x), and sulphur d ioxide (SO_2) are interesting as relevant noxious gases. In addition, pilot injection engines emit dust (soot). The measurement of this exhaust gas component was not the object of this study.

Legal Conditions

The Federal Immission Protection Act and the TA Luft provide the legal basis for the limitation of these exhaust gas emissions. **Table 1** lists concrete emission limits of the revised TA Luft [3]. These values apply to plants with a thermal firing output of less than 3 MW and refer to a reference oxygen content of 5% in the exhaust gas. The TA Luft is employed as an administrative regulation under the Federal Immission Protection Act.
According to the Federal Immission Protection Act, a permit is needed for an agricultural biogas plant if, among other conditions, a thermal firing output of 1 MW, a storage volume of 2,500 m^3, or a daily throughput of more than 10 tonnes of “waste requiring no special monitoring” are reached or exceeded [4]. These plants are required to observe the limits provided by the TA Luft. However, those plants for which no permit is needed according to the Federal Immission Protection Act must also conform with certain technical requirements [1]. Thus, regulations such as the TA Luft can also be employed at the discretion of the authorities as evaluation standards in construction permit procedures for the operation of an agricultural biogas plant.

Goals

The question arises whether the given limits can be met in practice. In general, it has been assumed that in practice the limits were not met by biogas-driven small plants. However, this has neither been confirmed nor disproved by long-term measurements. This study was intended to answer the following questions: How high are the emissions of CO, NO_x, and SO_2 from biogas-driven combined heat- and power plants? Do the emission rates remain constant or do they vary over the course of the measurement

Table 1: Limit values according to TA Luft (recommendations of the Bavarian Environmental Protection Agency in brackets) for low power combustion engines

Kind of engine	Carbon monoxide CO [g m^{-3}]	Nitric oxides NO_x als NO_2 [g m^{-3}]	Sulphur dioxide SO_2 [g m^{-3}]
Pilot-injection engine	2.0 (2.0)	1.0 (1.5)	0.35 (0.35)
Spark-ignition gas engine	1.0 (1.0)	0.50 (0.50)	0.35 (0.35)

series? Do the biogas-operated plants currently used in agricultural practice meet the requirements of the TA Luft? Are the given limits appropriate for the current technical standards of small and very small plants? Are there differences in the emission behaviour of different output classes of combined heat- and power plants? Does gas quality influence emission behaviour?

Measuring Set-Up

In a pilot-injection engine, an igniter oil (e.g. heating oil) is injected into the compressed fuel gas. Due to the high compression, this mixture ignites by itself. The engine exhaust gases produced are measured in the chimney. **Figure 1** provides a schematic overview of the measuring arrangement. If the exhaust gases from several combined heat- and power plants were fed into a common chimney, only the plant to be examined was operated during the measurement in order to prevent the mixing of the exhaust gases. The operators of the plants were required to start the combined heat- and power plant at least one to two hours before the beginning of the measurements so that the engines had reached their operating temperature before the measurement began.

Measuring Schedule

In order to obtain a long-term overview of the emissions of the measured combined heat- and power plants, one measurement each was carried out in the summer and in the winter. In addition, two measurements each were taken in the transitional periods. Each series of measurements comprised five to seven single measurements per unit. One single measurement included the measurement data of a 20-30 minute period at 10-second intervals. The exhaust gas measurements were taken with the measuring instrument TESTO 350. This mobile exhaust gas analyzer measures the exhaust air components CO, NO_x, and SO_2, as well as other parameters shown in **Table 3**. In addition to the exhaust gas emissions, the methane content of the biogas to be burned was determined with the aid of the measuring instrument ANSYCO GA 94 (**table 4**). This mobile measuring unit allows CH_4, CO_2, and O_2 in the biogas to be determined.

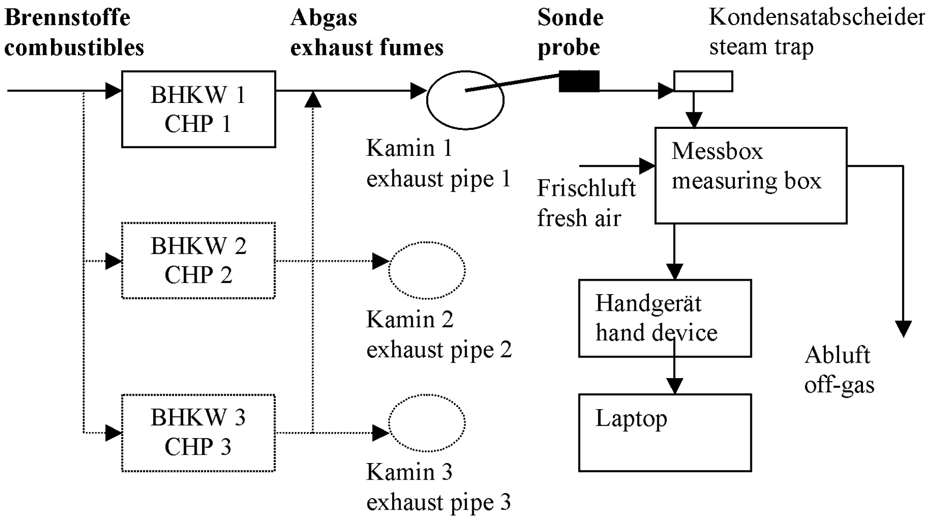

Figure 1: Schematic overview of the measurement set-up

Measurement Objects

The choice of the engines to be examined followed the results of a survey of Bavarian biogas plant operators [2]. The distribution of the electric power of the combined heat- and power modules of agricultural biogas plants covered in this survey ranges up to a maximum of 160 kW. Most engines reach an electric power of 10 kW to 50 kW. Many units can also be found in the 50 kW to 80 kW range. Four pilot-injection units with different power were chosen (**table 2**). In practice, pilot-injection engines are used more often than spark-ignition gas engines [2]. Moreover, the emission behaviour of pilot-injection engines is significantly more unfavourable. In addition to the pilot-injection units, a spark-ignition gas engine of the lowest power class (13 $kW_{el.}$) was covered in the study in order to gain insights into the emission behaviour of a very small unit common in practice. Since the beginning of the measurements, this engine has been a back-up unit which was started up exclusively for the measurements.

Table 2: Description of assayed engines according to power classes

Power [$kW_{el.}$]	Number of cylinders	Cubic capacity [l]	Represented power class [$kW_{el.}$]
22	3	2.9	<34
50	4	4.6	34-66
80	6	6.0	67-99
132	6	12.0	>99

Table 3: Means of emission measurements for each engine over the whole measuring period (GM = gas engine, ZS = pilot injection engine)

Measured variable	Unit	13 $kW_{el.}$ GM	22 $kW_{el.}$ ZS	50 $kW_{el.}$ ZS	80 $kW_{el.}$ ZS	132 $kW_{el.}$ ZS
CO	g m^{-3}	> 3.3	> 3.5	2.5	3.3	1.1
NO	g m^{-3}	0.63	0.71	1.09	0.30	1.05
NO_2	g m^{-3}	0.03	0.00	0.07	0.23	0.04
NO_x	g m^{-3}	0.99	1.09	1.73	0.68	1.66
SO_2	g m^{-3}	0.00	0.01	0.04	0.00	0.03
O_2	Vol.-%	4.5	4.1	9.2	10.5	8.9
CO_2	Vol.-%	15.7	14.3	10.2	8.8	10.2
Exhaust gas temperature	°C	51	255	215	182	323
qA	%	0.2	10.1	10.4	8.8	13.8
λ	-	1.27	1.26	1.81	2.05	1.78

Table 4: Composition of biogas (GM = gas engine, ZS = pilot injection engine)

Measurement series	Measured variable	Unit	13 kW GM	22 kW ZS	50 kW ZS	80 kW ZS	132 kW ZS
1 Sommer	CH_4	Vol.-%	47	68	52	60	59
	CO_2	Vol.-%	38	31	42	40	39
	O_2	Vol.-%	1.4	1.0	0.4	0.5	0.1
2 Autumn 1	CH_4	Vol.-%	50	k.A.	55	58	k.A.
	CO_2	Vol.-%	39	k.A.	38	k.A.	k.A.
	O_2	Vol.-%	2.5	k.A.	0.3	k.A.	k.A.
3 Autumn 2	CH_4	Vol.-%	k.A.	63	55	67	57
	CO_2	Vol.-%	k.A.	37	39	33	43
	O_2	Vol.-%	k.A.	0.4	1.5	0.4	0.1
4 Winter	CH_4	Vol.-%	49	61	54	63	50
	CO_2	Vol.-%	43	35	38	35	47
	O_2	Vol.-%	0.9	0.1	1.5	1.5	0.1

Data Processing

The measurement values of the exhaust gas analyzer stabilize only after a certain time when concentration in the measuring unit has adapted to the concentration of the sample air. This applies in particular to the CO values. In order to exclude these start-up values, which are obviously too low, from the calculations, they were removed from the data material. The measurement values were converted to match a reference oxygen content of 5 % like the limits stated under point 5.4.1.4 of the TA Luft. If the shut-off limit of the measuring instrument (3,000 ppm) was exceeded during a CO measurement, this limit was considered the minimum emission value. Statistical tests were carried out based on half-hour means. The data were evaluated using the statistics program package SPSS 10.0 for Windows®.

Results

Table 3 provides an overview of all measured parameters of the investigated engines over the whole measuring period as averaged mean values of a measurement series. For the 13 $kW_{el.}$ spark-ignition gas engine and the 22 $kW_{el.}$ pilot-ignition engine, it can be assumed that the CO emission value is higher than the stated mean value because these engines exceeded the measuring range of the instrument at least during certain periods. Table 3 and Figure 5 show that none of the engines examined fully meets the emission limits listed in Table 1. At a total average of more than 3.3 g m^{-3} CO and 0,99 g m^{-3} NO_x, the measurement results of the additionally measured 13 $kW_{el.}$ spark-ignition gas engine exceed the stated TA Luft limits for biogas-driven combustion engines with spark ignition (table 1). Depending on the plant and the series of measurements, the oxygen content ranged from 4.1 to 10.5 vol.-%. The CO_2 content, which ran counter to the oxygen content, amounted to 8.8 to 15.7 vol.-%. Analogous to the oxygen content, the λ value also ranged between 1.3 and 2.1. The exhaust gas temperatures were very different from plant to plant and ranged from 51°C to more than 320°C. As a result of the higher exhaust gas temperature, exhaust gas losses were at a higher level as well.

The mean CO emissions of the individual series of measurements ranged between 0.7 g m^{-3} in the largest unit and more than 4.2 g m^{-3} in an extreme case during partial load operation of the 80 $kW_{el.}$ engine (**figure 2**). At average values of 0.6 g m^{-3} to 1.9 g m^{-3}, nitric oxide emissions ran counter to the CO emissions of the individual engine (**figure 3**). In addition, Table 3 shows that the largest part of the NO_x emission is released in the form of NO. This must mainly be explained as a result of the high combustion temperature. After NO leaves the chimney, however, it is oxidized into the more harmful NO_2. Therefore, NO_x values are usually indicated in terms of NO_2 concentration values. A significant release of SO_2 could not be proven with the aid of the measuring systems used (measuring range 0 to 5,000 ppm, measuring accuracy <5 ppm at values below 100 ppm). The total average of the measured SO_2 emissions reached its peak at the 0.04 g m^{-3} emitted by the 50 kW engine and thus ranged far below the limit of 0.35 g m^{-3} required by the TA Luft.

Furthermore, the measurement values also indicated that engine setting and maintenance condition exerted a positive or a negative influence on the emission behaviour of the engine. Thus, the slow loosening of an engine bolt due to increased gas pressure and the tightening of this bolt were reflected by the exhaust gas measurements. **Figure 4** illustrates this behaviour using the temporal course of the NO_x and O_2 measurement values. During this malfunction, the CO values increased above the shut-off limit of the CO measuring cells. Therefore, a quantitative assessment cannot be given here. A spark-plug change in the 13 $kW_{el.}$ spark-ignition gas engine after the third series of measurements resulted in a considerable reduction in CO emissions from 3.5 g m^{-3} before to 2.5 g m^{-3} (not shown).

In Figure 2, a systematic influence of the outdoor temperature represented by the

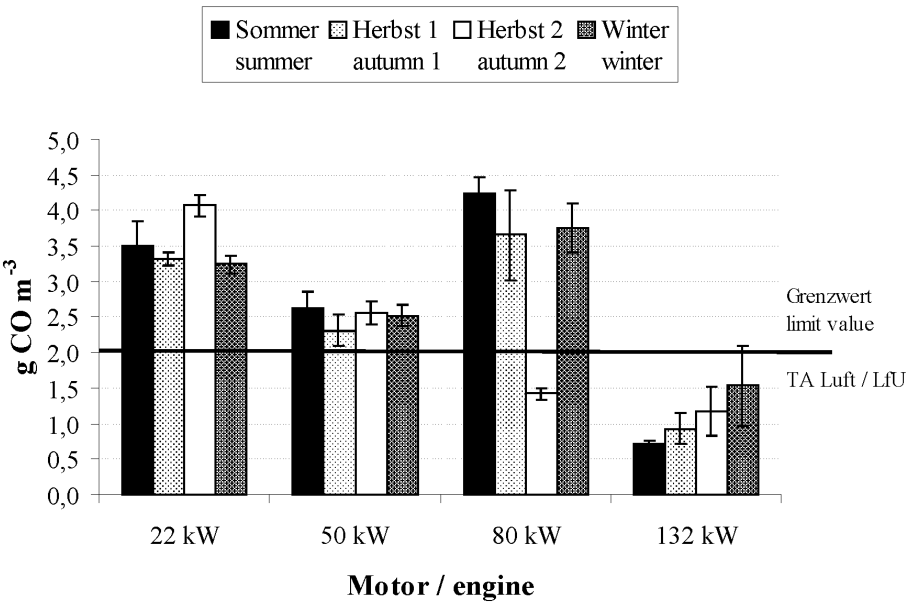

Figure 2: Mean carbon monoxide emissions of the investigated engines (with standard deviations of the CO-emissions during each measuring period)

measurement series cannot be detected. This can be attributed to the fact that the engine usually aspirates the combustion air from the warm room where it is installed. CO concentrations fluctuated in a more or less wide range. Only in the 132 $kW_{el.}$ engine did the values exhibit a continuous increase over the course of the measurement series. The assumed reason for this increase is a reduction in the methane content. This point will be discussed below.

Analysis and Discussion

Figure 5 shows that none of the investigated engines meets both the CO and the NO_x limits of the TA Luft. The significance of deviations from the limit in the case of values exceeding the limit or ranging below it was examined statistically and is extremely significant ($p < 0.001$) except for the nitric oxide emissions of the 22 $kW_{el.}$ engine, where the measurement values are within the limit range.

If the engines are compared to each other with regard to emissions, they exhibit extremely significant differences ($p < 0.001$) at a global test level for both the CO and the NO_x values. This is confirmed by a multiple mean value comparison. Only the pairwise comparison of the CO emission of the 22 $kW_{el.}$ engine and the 80 $kW_{el.}$ engine ($p = 0.37$) and the NO emissions of the 50 $kW_{el.}$ and the 132 $kW_{el.}$ unit ($p = 0.93$) do not allow any significant difference to be discerned (cf. figure 5).

A comparison of the measurement series of one engine also enabled extremely significant differences to be established ($p < 0.001$). Only the CO values of the 50 $kW_{el.}$ engine and the NO_x values of the 132 $kW_{el.}$ unit cannot reach this significance level (cf. figure 2 and figure 3). The difference between the series of measurements allows the conclusion to be drawn that one single measuring date does not provide meaningful results for the evaluation of the emission behaviour of an engine in practical operation because the values may deviate from those of another measuring day due to varying conditions. This is particularly interesting if the emission level of a measured engine ranges near the limits. These deviations may be caused by different factors, such as changed engine settings, maintenance work, a lack of gas, or variations in the methane content of the biogas. Therefore, an investigation should extend over several days so that a meaningful overview of an engine´s emission behaviour in practical operation can be gained beyond the fluctuation range of gas quality.

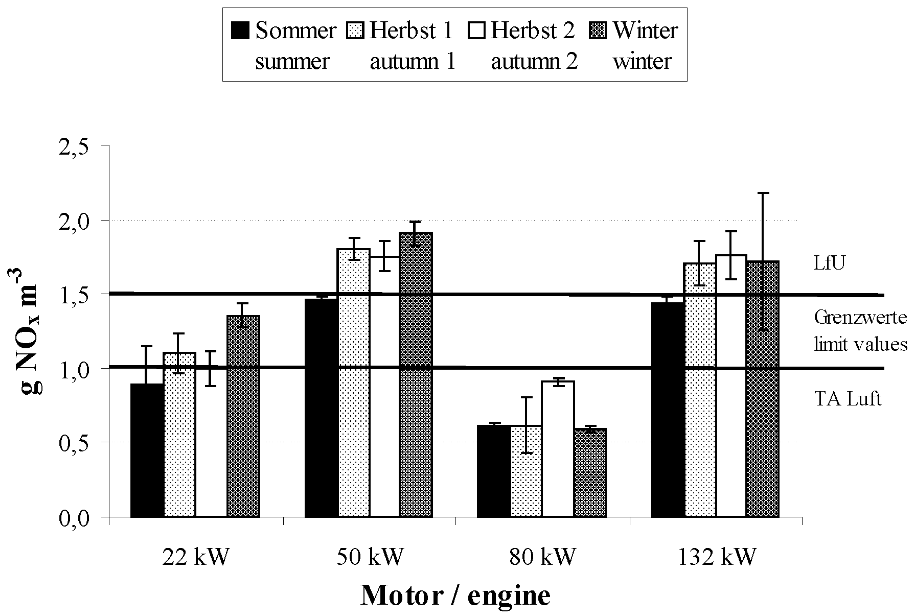

Figure 3: Mean nitric oxide emissions of the investigated engines (with standard deviations of the NO_x-emissions during each measuring period)

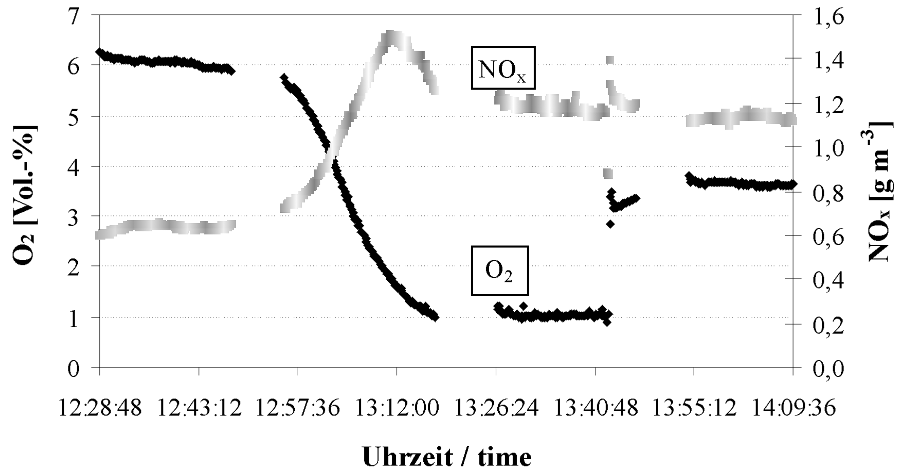

Figure 4: Measured O_2 and NO_x emissions during an engine malfunction

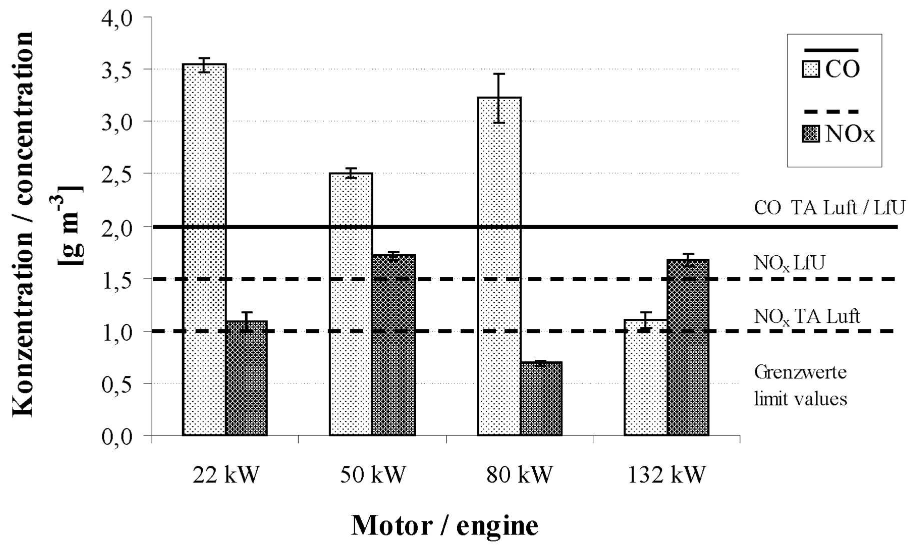

Figure 5: Mean emissions based on half-hour-means, standard error, TA Luft limit values (CO: 2.0 g m^{-3}; NO_x: 1.0 g m^{-3}) and recommendations of the Bavarian Environmental Protection Agency (CO: 2.0 g m^{-3}; NO_x:1.5 g m^{-3})

It is remarkable that CO emissions decrease with growing engine power (figure 2). This can be seen very clearly in the third series of measurements in particular (autumn 2). Apart from the exceptional values of the 80 $kW_{el.}$ unit, this trend can also be observed in the other series of measurements. The increased emissions of this engine, which obviously run counter to the trend at first glance can be attributed to the fact that due to a lack of gas the engine had to operate under partial load on these measuring days. Hence, this engine exhibits relatively high CO- and relatively low NO_x values in the exhaust gas due to incomplete combustion and lower combustion temperatures. Even in the case of this engine, however, the trend towards a decrease in CO emissions with growing unit size was confirmed when the engine was measured under full load in measurement series 3. The decreasing tendency ($r = -0.66$, $p < 0.001$, incl. values from the partial load operation of the 80 kW unit) may be caused by the larger cubic capacity of the engine cylinders allowing for more complete biogas combustion. Larger engines are possibly also maintained more carefully.

During full-load operation of the 80 $kW_{el.}$ unit, the standard deviations of the measurement values of the third measurement series (autumn 2) were considerably lower than in the other series of measurements (figure 2). This must obviously be attributed to the more even combustion behaviour of the engine under full load in contrast to partial load operation. This example shows that changing combustion conditions exert a strong influence on emission behaviour and that optimal engine setting with regard to the fuel-/air ratio provides great emission reduction potential.

When considering the CO and NO_x values, one notices that a high CO emission value expectedly corresponds with lower NO_x emissions and vice versa. This is caused by higher temperatures and complete combustion. In this case, CO emissions sink. However, the formation of NO is strongly dependent upon the temperature and is thus favoured. The 132 $kW_{el.}$ unit, for example, illustrates this effect: this unit has the lowest CO emissions, the highest exhaust gas temperatures, and relatively high NO_x values. The extremely low SO_2 values of the measurements carried out (max. 0.04 g m^{-3}) must be considered with reservations because the biogas to be burned may contain significant quantities of hydrogen sulphide. Hydrogen sulphide in biogas was not measured in this study. In a plant which features a built-in biogas analyzer, hydrogen sulphide concentrations between 200 ppm and 600 ppm were measured even though the gas was desulphurized. At these values, SO_2 contents of ca. 0.06-0.18 g m^{-3} would have to be expected in the uncleaned exhaust gas. The following assumptions can be made about the whereabouts of the sulphur: one part gets into the engine oil circuit, acts aggressively there, and is responsible for the corrosion of the fittings and the engine [1]. Another part of the sulphur is released into the atmosphere with the exhaust gas (possibly as SO_3 or H_2SO_4), or it is dissolved in the condensate water and retained over the heat exchangers or discharged from the measuring system. Hence, the possibility exists that the actual SO_2 concentrations could not be measured. Due to the desulphurization of the biogas, which is carried out everywhere, the SO_2 content of the exhaust gas is probably very low.

The increase in CO emissions over the course of the measuring period of the 132 $kW_{el.}$ unit (figure 2) can be explained as a result of the deterioration in combustion quality due to the sinking methane content of the biogas [5]. An agitator defect in the fermenter could be a possible reason for the sinking methane content. This results in a lower degree of homogenization of the fermentation substrate so that the conditions for methane production are no longer optimal.

Stationary exhaust gas measurements in another pilot-injection unit (160 $kW_{el.}$) confirmed the effect of increased CO emissions combined with sinking methane contents of the biogas. For this purpose, the engine was run with unaltered biogas (methane content ca. 60%) and biogas which was diluted down to a methane content of ca. 40 to 50% using CO_2 in a mobile gas store. On average, the TA Luft emission limit of 1.0 g m^{-3} for NO_x was just met at the lowest methane content. In the two other cases, it was exceeded (table 5). With decreasing methane content of the biogas, NO_x concentrations in the exhaust gas diminished, while CO- and methane emissions (“methane slip”) grew, which can be explained as a result of slower, colder and, hence, more incomplete combustion (**figure 6**). However, all CO values ranged significantly below the relevant TA Luft emission limit.

On average, the measurement values of formaldehyde as a product of incomplete methane combustion exceeded the TA Luft limit in all three operating states. The highest formaldehyde concentrations in the exhaust gas occurred at the lowest methane concentration in the fuel gas, which can be considered in accord with the measurement values of total C and methane in the exhaust gas (**table 5**). In this process, formaldehyde may be produced through postoxidation of methane in the exhaust manifold. The measured exhaust gas concentrations of butene, pentene, acetone, benzene, and toluol all ranged only slightly above the detection limit of 0.1 mg m^{-3} (data not shown).

The measurement results indicate that, if possible, the setting of the engines should be adjusted to the methane content of the biogas. However, this is problematical because biogas quality often fluctuates in practice.

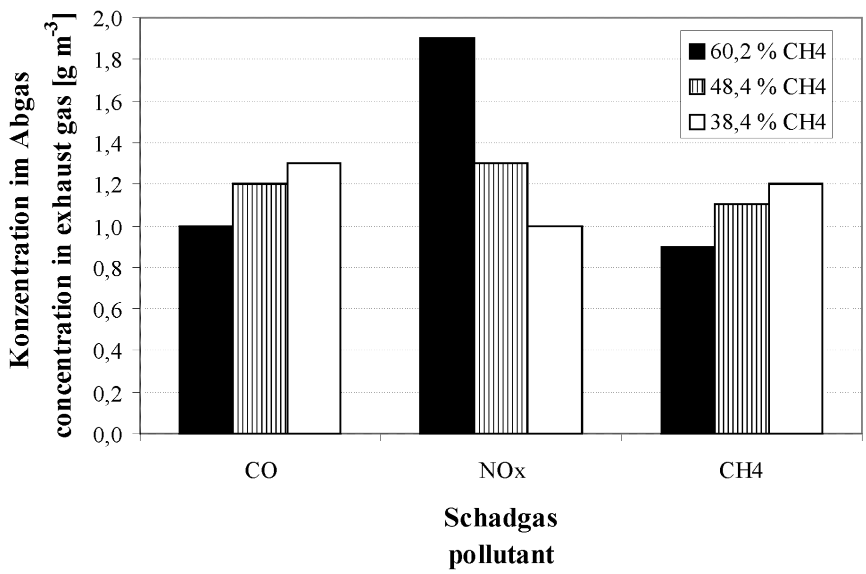

Figure 6: Mean exhaust gas concentrations at varying methane concentrations in the biogas

Table 5: Means of biogas- and emission measurements for a 160 $kW_{el.}$ engine

Measured variable	Unit	Average of the measurement values		
Methane in the biogas	Vol.-%	60.2	48.4	38.4
Oxygen	Vol.-%	7.5	7.4	7.2
Carbon dioxide	Vol.-%	14.1	16.8	18.3
Carbon monoxide	g m^{-3}	1.0	1.2	1.3
Nitric oxides (as NO_2)	g m^{-3}	1.9	1.3	1.0
Sulphur oxides (as SO_2)	g m^{-3}	0.06	0.06	0.05
Total C	mg m^{-3}	1016	1329	1500
Methane in the exhaust gas	mg m^{-3}	898	1104	1218
Total dust	mg m^{-3}	8.4	4.9	4.9
Formaldehyde	mg m^{-3}	111	87	131

Conclusions

The assumption that existing small plants do not fully observe the TA Luft limits in practice was confirmed by the investigation of pilot-injection engines. Due to the short service life of catalyzers in combined agricultural heat- and power plant modules, the use of a catalyzer for exhaust gas aftertreatment is not yet state of the art. Given the current technical standard of small plants, the strict application of the TA Luft should be avoided in construction permit procedures at the discretion of the licensing authority for the sake of the promotion of environmentally friendly energy generation. Instead, emission limits more appropriate for practice like those recommended by the Bavarian Environmental Protection Agency ([6], table 1) should be employed. Since the emissions of an engine are influenced by varying conditions (e.g. course of the process and, hence, gas quality, gas quantity, engine setting, maintenance condition) in practical operation, more frequent measurements may be required for the emission behaviour of a combined heat- and power plant to be evaluated with sufficient accuracy with regard to optimal engine setting. In addition, statistical tests have shown that there was a significant difference between the exhaust gas values of different engines with a tendency towards lower emissions in larger units. With regard to emissions, the current trend towards more powerful combined heat- and power plant modules [2] must therefore be assessed positively. Regular, proper maintenance of the engines, which has so far hardly been carried out in practice, could also help meet the emission limits. Examples of maintenance measures are a regular change of injection nozzles and optimized engine settings with regard to individual biogas quality.

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The studies were financed by the Bavarian State Ministry of Rural Development and the Environment and scientifically accompanied by the Bavarian Environmental Protection Agency.

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