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## Investigation of methane losses during the caloric upgrading of biogas for grid injection

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## **1 Introduction:**

In the last years a large number of bio gas plants were build which generate gas mixtures of methane and carbon dioxide usually up to 1000 m<sup>3</sup>/h in a single plant. Often this gas is used as fuel for electrically power generation but there is also an interest to inject the gas into the public gas grid. For this purpose a purification is needed which increases (upgrades) the methane partition of the injected gas in order to realize gross caloric value similar to the natural gas in the grid. An important issue of CO<sub>2</sub> - separation is the emission of methane into the atmosphere by losses which is dependent from the applied technology.

In order to investigate bio gas plants for gas injection in the public grid in Germany a joint research project called "MONA" was established <http://www.mona-biomethan.de/>. The overall objective of this project is an evaluation of biogas upgrading technologies for biogas grid for injection or utilization as vehicle fuel by an integral view that means with regard to environmental impact, economy, operation and technology.

In this project - plants already in operation - shall be monitored. The PTB part in this project is the measurement of the methane losses via exhaust gas of the biogas upgrading facilities and eventually after the exhaust gas treatment.

## **2 Biogas upgrading technologies**

For the upgrading of raw biogas there are four steps generally needed:

- Desulphurization
- Drying
- CO<sub>2</sub> extraction
- Optional: extraction of other contaminates

Depending on the upgrading technique the order of these process steps may differ.

The most common upgrade technologies are the pressure swing adsorption, water scrubbing, chemical scrubbing and membrane technologies [1].

The pressure swing adsorption uses activated carbon or zeolites to extract the CO<sub>2</sub> gas component. To regenerate the adsorbent the pressure is decreased. Normally several vessels stand together for continuous operation.

Another upgrading process is water scrubbing. The carbon dioxide dissolves in the water. The technology uses the effect, that methane is dissolved to a much lower extent. Dissolved methane is recovered in flash tank to minimize the methane loss by depressurization. The water is regenerated in a desorption column.

In a chemical scrubbing process the carbon dioxide is bound chemically. The advantage of chemical scrubbing is the low methane loss. To bind the carbon dioxide a scrubbing liquid is used which consists of water and amine for example. The regeneration of the scrubbing liquid is carried out by heating it in separate vessels.

The membrane technology is the least common of these examples. It uses the different diffusion rate of methane and CO<sub>2</sub>. Solid membranes can be constructed as e.g. hollow fiber modules, which give a large membrane surface per volume and hence compact units. Solid membranes can be constructed from several polymers as e.g. acetate-cellulose. [2]

The carbon dioxide extraction technology has an influence on exhaust gas composition. Especially in the exhaust of the scrubber plants moisture up to 100% is expected. Depending on the quality of the extraction process some plants need a so called "exhaust after treatment" where methane residues are removed from the gas usually by burning.

## **3 Determination of methane loss**

There are two ways methane gets emitted in a biogas upgrading facility:

- Fugitive emissions by leakage, maintenance, discharge via process chemicals, emissions in case of an emergency,
- Discharge via exhaust gas of the biogas upgrading facilities and eventually after the exhaust gas treatment.

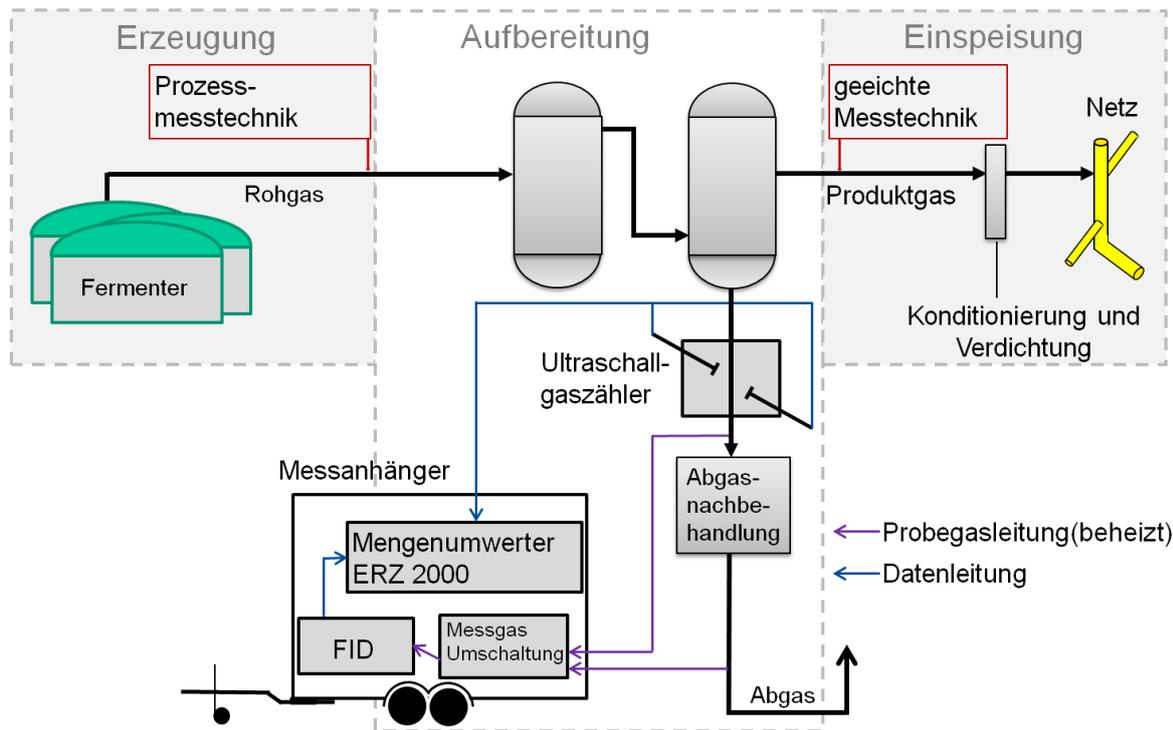
Concerning the measurement of emissions the project is focused on the exhaust gas investigation.

The maximum emission of methane in the exhaust gas of the upgrading facility is limited by 0.2% [3] of the total amount. The different upgrading techniques provide in general lower concentrations during continuously run.

In order to draw a balance over the upgrading facility it is necessary measure at least two of the three gas flows (see figure 1). At the input of the upgrading facility a flow rate measurement device as well as gas composition analyzer is installed. But there is no reliable information available about the uncertainty of these process measuring devices. At the injection

point in the grid flow meters and process gas chromatographs are installed, which are approved and calibrated. These devices provide an uncertainty below  $U = 1\%$ .

Even if the process measurement devices would provide an uncertainty of  $U = 1\%$  the balance does not allow a significant conclusion on the concentration of methane.



**Fig. 1: General arrangement for the exhaust gas investigation of biogas**

Hence, in order to benchmark the upgrading techniques it is necessary to measure the exhaust gas flow rate and the concentration of methane in the exhaust gas simultaneously.

The volume flow rate measurement is performed by an ultrasonic gas meter designed for this purpose. The advantage is a low pressure loss. In order to use the USM it is foreseen to drill holes in the exhaust pipe to gain access to the flow. Further details are explained in chapter 4.3. After the end of a campaign the holes will be closed by a single commercial available stainless steel sleeve. The uncertainty of the flow rate measurement depends on the flow profile on the place of installation and hence on the available straight forward input length in front of the meter.

The quantification of the total amount of exhaust gas is done by revaluing the flow rate at operating state to standard state by a commercially available conversion device and by measuring the concentration of methane by an online “flame ionization detector”. The detector uses hydrogen and synthetic air provided from bottles for the detection flame to avoid environmental influences on the results.

In the project it is intended to measure for two weeks at several plants. To be mobile all equipment is

installed and stored in a trailer. The conversion device provides also data storage and a remote reading of data. This is necessary to allow an automatic run. Only at the beginning and the end of the 2 week campaign personnel need to be on site.

## 4 Measuring equipment

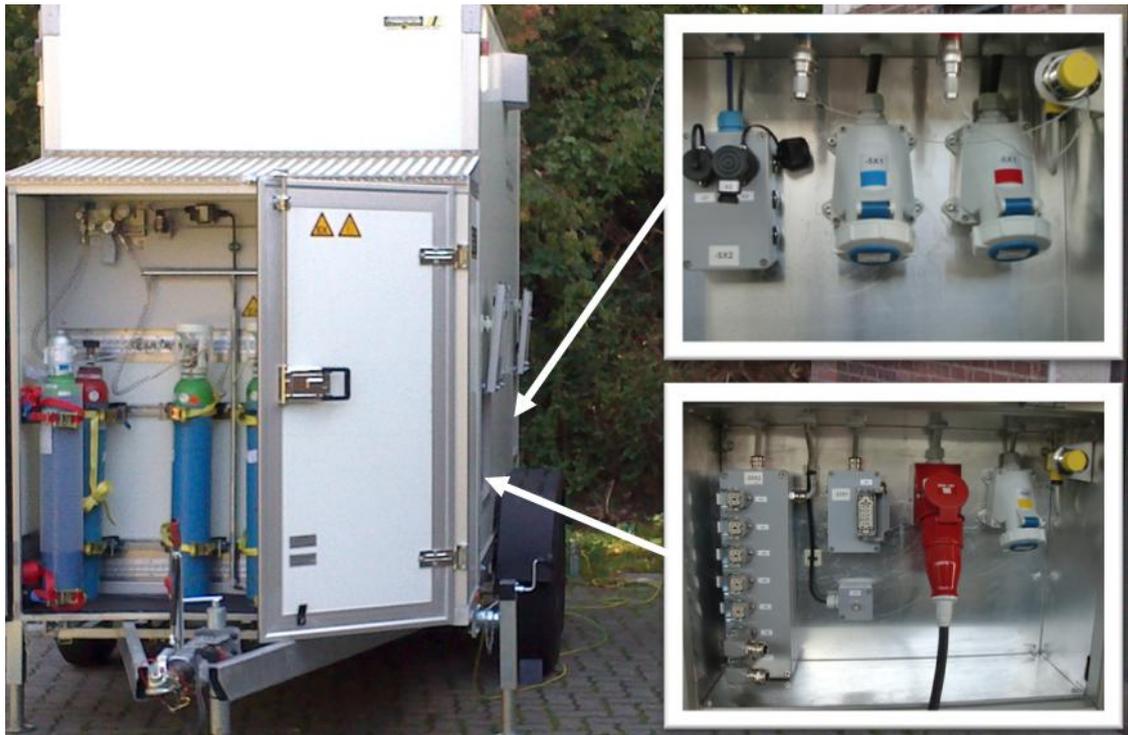
### 4.1 Measurement trailer

In order to investigate different plants a trailer was engineered and constructed by RMG<sup>1</sup> in cooperation with PTB. The following figure 2 shows the front and side view of the Trailer. The trailer is designed to stand alone for two weeks. The only needed supply is polyphase electricity of 16 A. The design considers protection against access of unwanted animals like vermin from the outside. Because of the use of explosive gases a safety concept for the inside was established. The safety concept consist of a gas detector, an explosion protected fan, uninterruptible power supply and a temperature sensor for the inside. This concept allows an automatic emergency shutdown of the trailer systems.

<sup>1</sup> RMG Messtechnik GmbH, Otto-Hahn-Straße 5, D-35510 Butzbach, Deutschland



**Fig. 2: Photograph of the measurement trailer**



**Fig. 3: Photograph of front view of the measuring trailer**

Inside is enough space for the complete equipment such as the heated pipes for the gas sampling, the ultrasonic metering modules, the data cables etc.. In a separated room in the front the gas storage for combustion gas and combustion air is located. This storage is connected with pipes to the flame ionization detector. It offers space for up to seven 10-Liter gas bottles. So the calibration gas can also be stored there. In the main room the installation is

carried out. A sample gas switcher, a filter and the gas pump to carry the sample gas to the flame ionization detector are arranged on one wall of the trailer. To avoid condensation most of the gas containing components are actively heated. A 19 inch standard rack serves as control cabinet. Beside the flame ionization detector and safety system the controllers for all electric components, the remote data exchange system and the volume conversion

device are installed in the rack. The volume conversion device collects all data and is used as a data logger too. The logging interval is one minute. As conversion device an approved device for custody transfer of company RMG, type ERZ 2000 is chosen because there is a remote reading and data bank software available for the data analysis. Via the remote data exchange system it is possible to have a remote online view on the collected data. Each day the electronic data interchange sends a package of data to a computer used for supervision at PTB. This package consists of the information collected since the last call.

## 4.2 Methane measurement

The methane concentration measurement is based on the flame ionization principle. One of the main advantages of using a flame ionization detector is its low sensitivity to moisture. An automatic process measurement device of company Siemens, type Fidamat 6 was chosen. It offers several detection ranges from 0 ... 10 ppm up to 0 ... 100000 ppm. The detection unit is temperature stabilized at 200°C. The system is able to start the detection flame automatically and switch of all supplies in case of a permanent failure.

For calibration gas is utilized with a certification from the Deutscher Kalibrier Dienst (German calibration service). Depending on the upgrading technique different concentrations are used to calibrate at the beginning of the campaign. It is planned to check the drift by using the calibrations gas during the campaign. The calibration gasses cover from 0.15 Vol.-% up to 5 Vol.-% methane in nitrogen matrix.

The methane emission will be calculated by the following formula:

$$E_{R,CH_4} = \int_t Q_{Exhaust} \cdot c_{m,CH_4} \cdot dt \quad (1)$$

$$c_{m,CH_4} = \frac{x_{V,CH_4} \cdot M}{V_{m,n}} \quad (2)$$

$t =$  two weeks

symbol	description	unit
$c_m$	mass concentration	g/m <sup>3</sup>
$M$	molare mass	kg/kmol
$Q_N$	flow rate	m <sup>3</sup> /h
$V_{m,n}$	molare volume at standard conditions	m <sup>3</sup> /kmol
$E_R$	emission rate at standard condition	mg/h
$x_{V,CH_4}$	volumetric concentration	-

## 4.3 Flow device

At the CO<sub>2</sub> exhaust of biogas upgrading facilities there is usually no need to measure the flow rate. Hence the piping does not allow installing a standard flow meter. Further considerable changes in the facility piping have to be checked by the authorities responsible for the work admission. Consequently a flow meter system is needed, that gears just a little into the structure. On the other hand a flow meter providing a sufficiently low uncertainty should come in use. In particular a low sensitivity to flow perturbations was of interest. Taking these preconditions in account a 1 path ultrasonic flow meter was considered as the most appropriate solution.

Together with PTB company SICK<sup>2</sup> has designed the ultrasonic flow meter module. It is based on the type Flowsic 600 for custody transfer which is able to measure under tough conditions. The design of the ultrasonic gas meter is in respect to the path lay out is similar to a clamp on system. As the gas is expected to have a high concentration of carbon dioxide and a pressure roughly at atmospheric conditions, the known clamp on systems probably will not provide sufficient signal levels. Therefore the exhaust pipe needs to have 2 holes at two opposite positions in respect to the length axis of the pipe. This allows to have direct contact of the ultrasonic transmitters with the gas and to have direct view from sensor to sensor.

The chosen meter is build for a gas speed range from 0.3 to 20 m/s and for diameters between 80 mm and 300 mm.

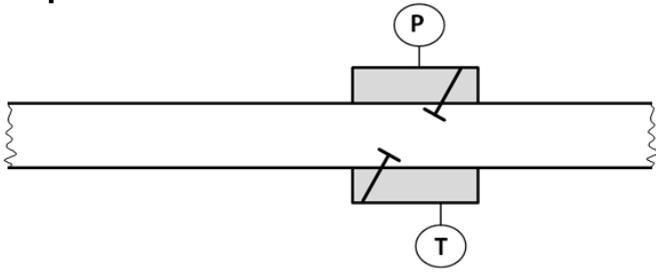
The module was designed as a sleeve that has to be fixed on the pipe. The sleeve has the advantage that it can be fixed on any location of the pipe as long as the diameter differs not too much from the nominal outer diameter ( $d_{out} = 89 \pm 2$  mm) DN80. For some plants the module may be installed as an additionally pipe (see figure 4 option 2). To install the sleeve, only two 28 mm drill holes are necessary.

During application the sleeve is fastened with stainless steel straps and pressed with its shoulders (orange mark in figure 6) at one site on the pipe. A silicone foam plate is stucked into both half shells of the sleeve in order to seal to ambient.

The sleeve has mechanical measures to allow a reproducible assembling. Hence ultrasonic path length and angle is always well defined.

<sup>2</sup> SICK AG, Erwin-Sick-Str. 1, 79183 Waldkirch, Germany

Option 1



Option 2

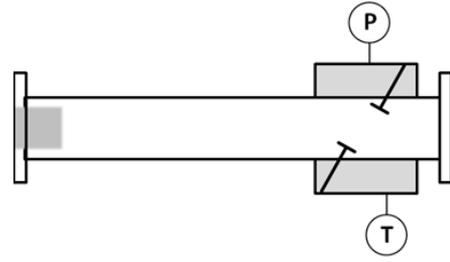
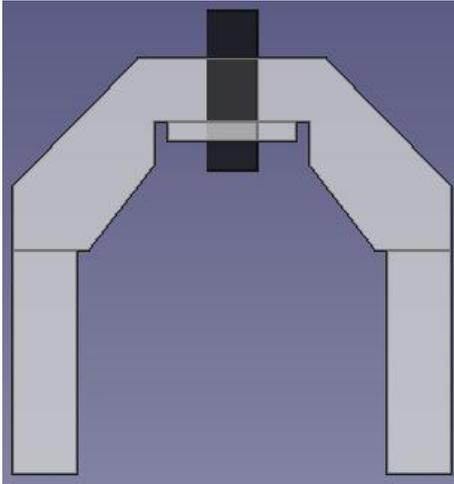
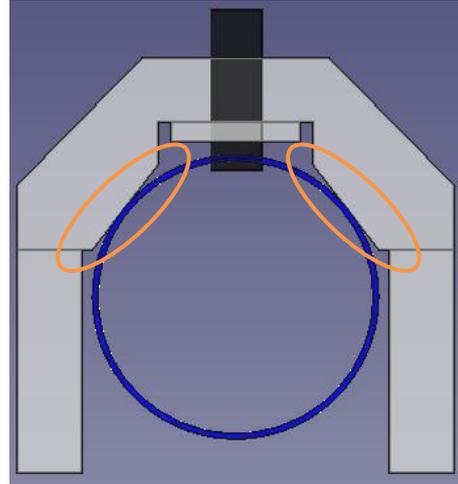


Fig. 4: Different use strategies

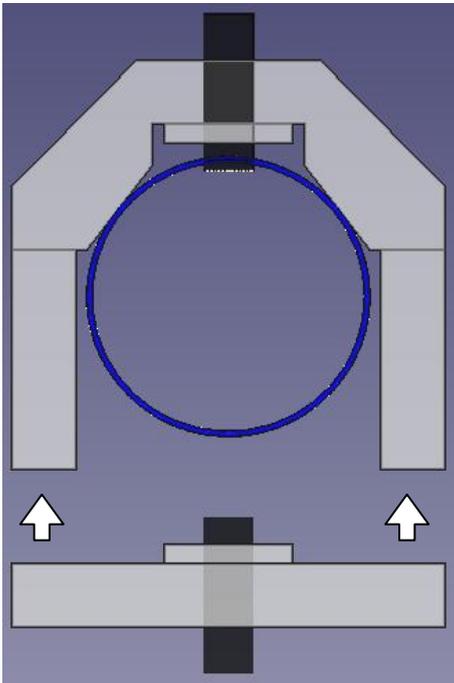
(1)



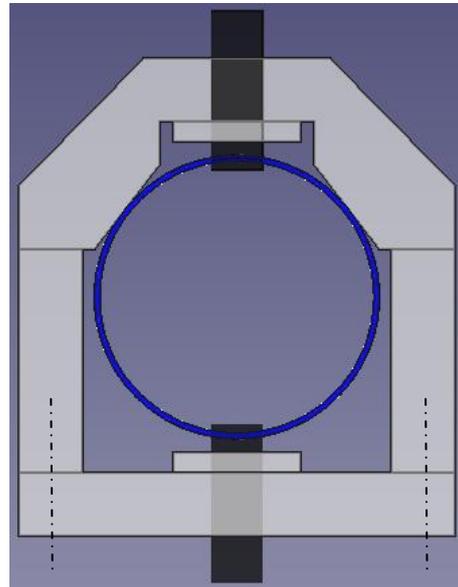
(2)



(3)

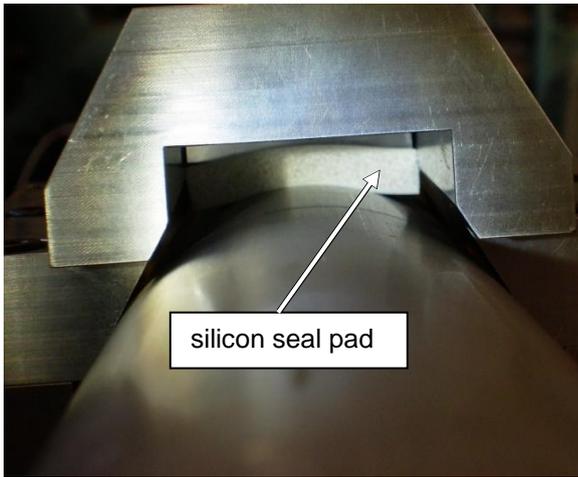


(4)



the lower part gets fastened by screws

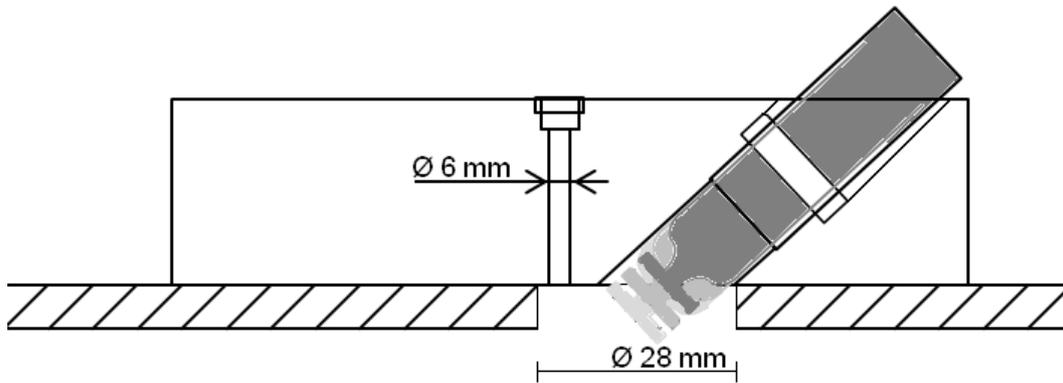
Fig. 5: Installation of the measuring sleeve



**Fig. 6: Photograph of the silicon foam pad for sealing the gap between sleeve and pipe around the transducer pockets**

As a special feature it is possible to vary the transducer position in the half shells of the sleeve as

figure 7 shows. There are three positions possible. In the deepest position the membrane of the transducer is nearly totally “wetted” by the flow (light grey). In the outer position (dark grey) the transducer does not wet the flow at all. In the outer position the membrane of the transducers is 8 mm pulled back. In addition a middle position is available where the membrane is 4 mm pulled back. As carbon dioxide absorbs ultrasonic waves quite severely the transducer frequency was chosen relatively low (135 kHz). The opportunity to pull the transducers into the flow allows it to reduce the distance and to have free propagation of the ultrasonic waves in order to reach sufficient signal levels for large pipe diameters. In case of a small pipe diameter the influence of the transducers on the flow may be reduced by pulling back the transducers into the sleeve. If not mentioned explicitly all tests were carried out with transducers pulled completely into the flow (light grey).



Abgasrohr

**Fig. 7: Sketch of a half shell of the sleeve.**

In order to measure pressure and temperature in each half shell of the sleeve a 6mm hole is realized.

The uncorrected volumetric flow rate  $Q_V^*$  is calculated from the mean velocity  $\bar{v}$  and the open cross-sectional area in the pipes measuring section:

$$Q_V^* = \bar{v} \cdot \frac{D_i^2 \cdot \pi}{4} \quad (3)$$

This intermediate result is dependent on Reynolds number and the properties of the flow profile (asymmetry, swirl). In order to correct the Re-influence by the following formula is usually applied:

$$Q_V = Q_V^* \cdot (1 + k(Q_V^*)) \quad (4)$$

The test results described in the following do not use a correction term.

$$k(Q_V^*) = 0 \quad (5)$$

symbol	description	unit
$D_i$	inner Diameter	m
$Q_V$	volumetric flow rate	m <sup>3</sup> /h
$\bar{v}$	mean velocity of the USM path	m/s

#### 4.3.1 Investigation of the characteristics of the flow metering unit

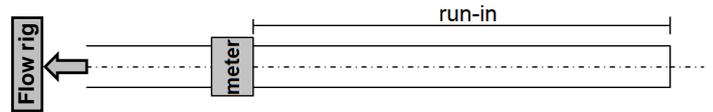
For the project an uncertainty of 2% for the volume flow rate determination is aspired. Therefore several tests were made to qualify the ultrasonic meter module.

In the first step the measuring characteristics under ambient air temperature were brought into focus. The

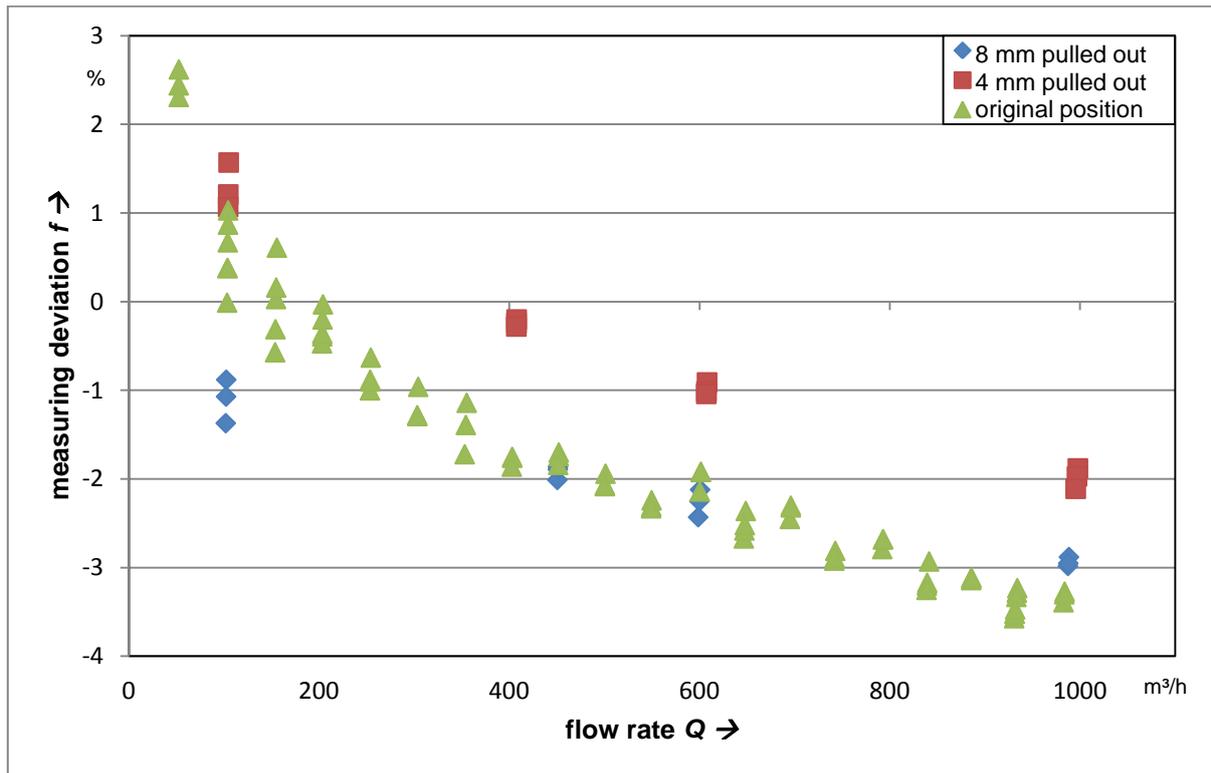
test was made at the nozzle test rig in the PTB. The flow medium was air. For the test ambient air temperature and gas temperature were at the same level. The temperature drifted not more than  $\pm 1^\circ\text{C}$  during one tests arrangement. For each test at one flow rate a measuring period of 100 seconds was applied.

Referring to figure 7 it is possible to rearrange the transducer position in the sleeve. This is done with

distance disks. The available disks are 4 and 8 mm. Thereby the three positions – full in flow, pulled back and out of flow – can be used.



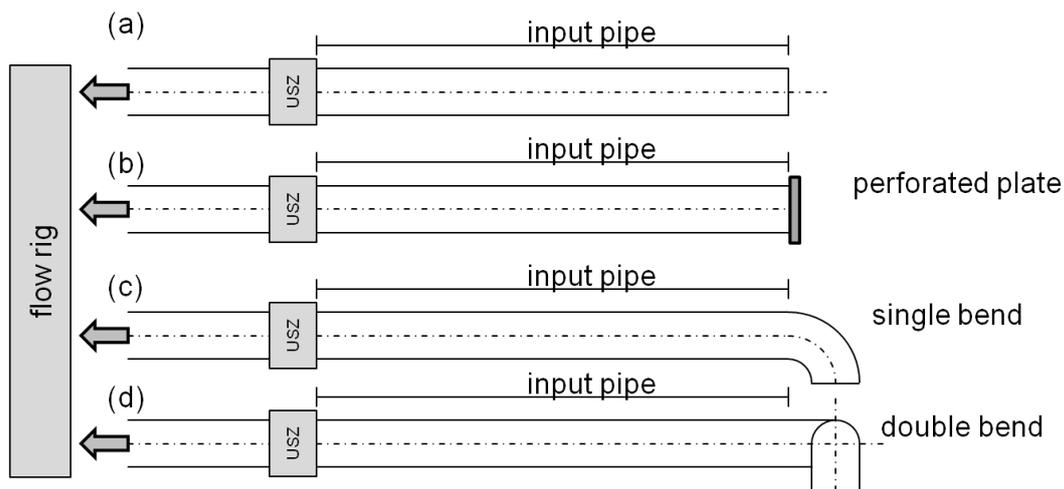
**Fig. 8: Test arrangement for different transducer depth**



**Fig. 9: Testing the different transducer positions**

The 4 and 8 millimeter position provide error curves which seem to be parallel while the deepest position leads to an increased flow rate dependency of the meter error at low flow rates. There were several measurements carried out after changing the transducer position. The reproducibility was inside  $\pm 0.5\%$ . This allows an correction after application of

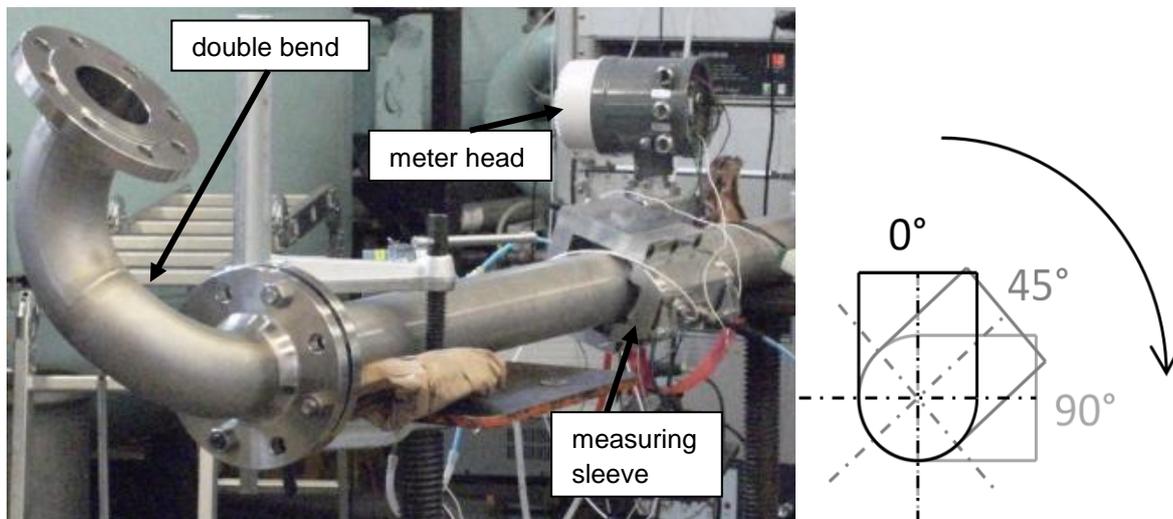
the module depending on the used transducer position. Concerning the input perturbations in a first step the investigations started by testing the module with a straight forward input pipe. Then there was added perforated plate (hole straightener), a bend and a double bend out of plane.



**Fig. 10: Test set-up with different upstream piping configurations**

The run in distance was 10D (figures 12 to 14) and 20D (figures 15-17). When testing with a bend the position of the bend was changed eight times, each time 45° turned clockwise. In the same way the double bend out of plane was tested but at 4 positions only. This high number of tests was made because the qualities of the results that can be reached with an one path ultrasonic system depend

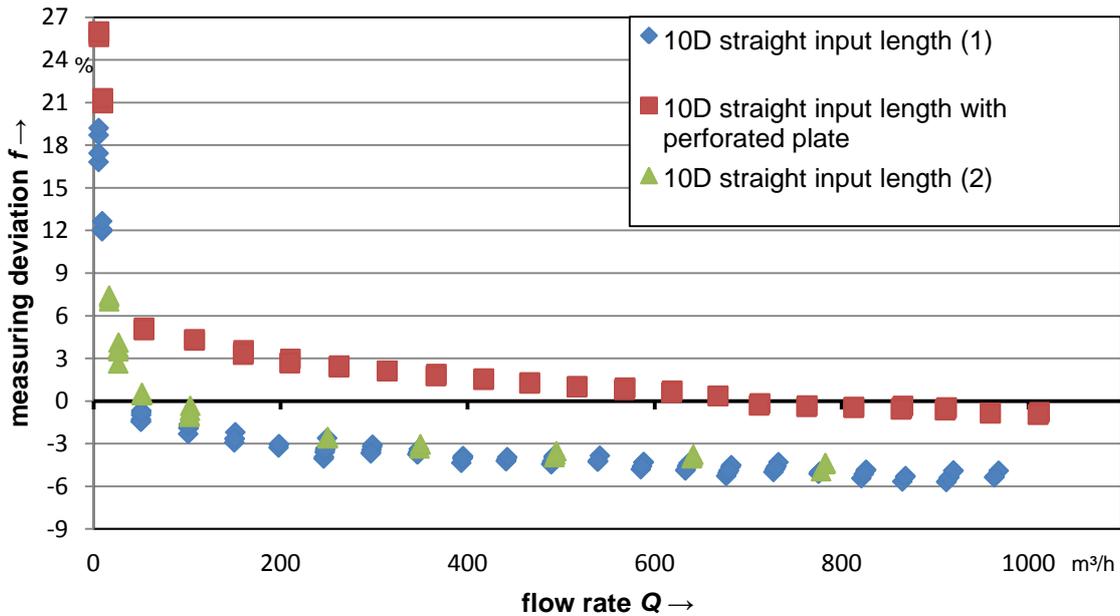
severely on the flow profile. Figure 11 shows a photograph of the installation and a schematic explanation of the investigated position of the test. The USM is path at 90° installed. The pipe size was DN 80. It is planned to a 6mm hole investigate 2 additional pipe sizes to check the upscaling behavior of the system. One of them will be DN 200.



**Fig. 11: Left: Photograph of a double bend out of plane at 315° position; right: schematic explanation of the investigated positions**

Each of the following charts presents the results of several test series. In each series several flow rates

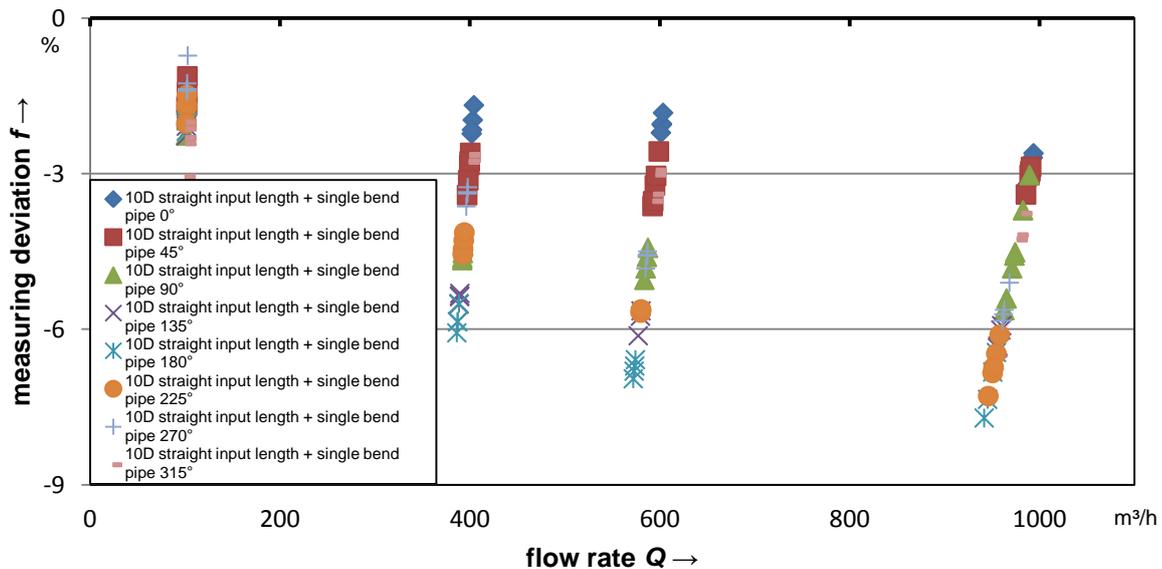
were tested. For each flow rate repeated tests were done.



**Fig. 12: Measuring series at 10D run in distance**

Figure 12 shows how much the upstream configurations affect the measuring results. The installation of a perforated plate led to an offset of

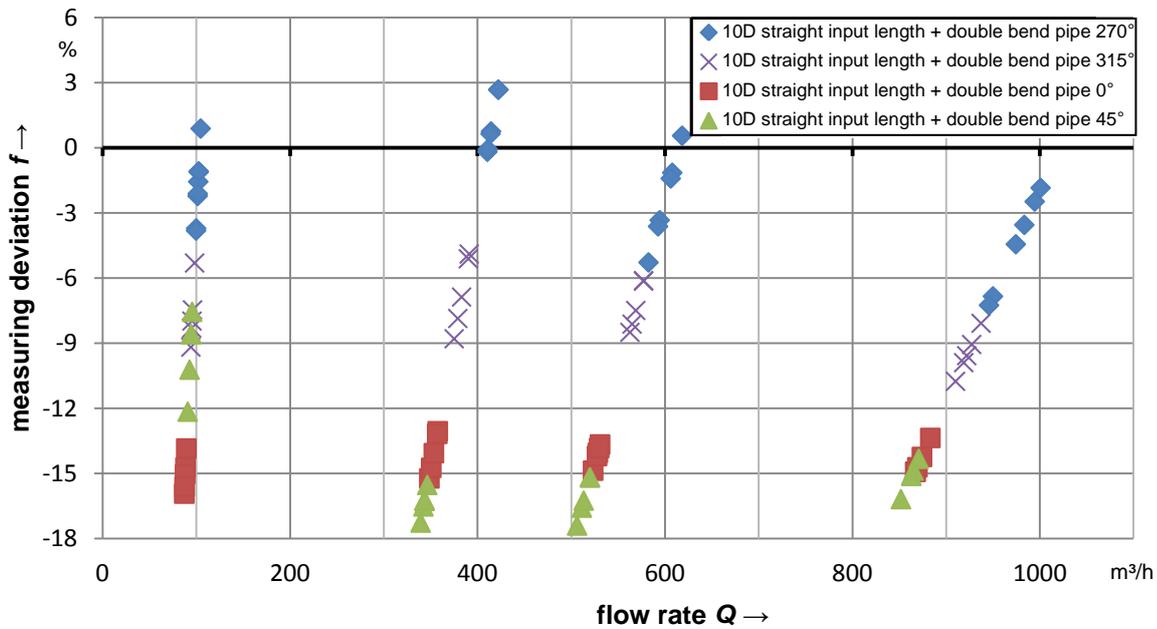
about 6% and reduced the scattering of the results in the whole flow rate range.



**Fig. 13: Measuring results with 10D straight pipe and different arrangement of a single bend**

The installation angle of the single bend relative to the position of ultrasonic path led to a drift of the measuring results. The repeatability of the results is independent from the installation angle of the bend.

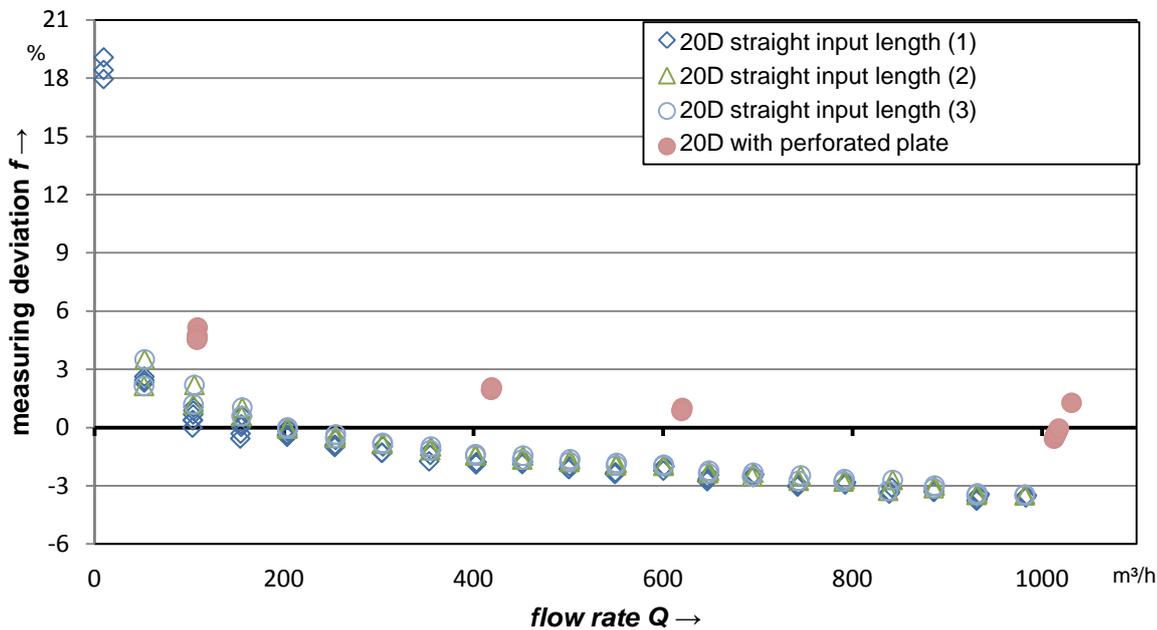
The influence of the bend is small at low flow rates ( $\pm 1.5\%$ ) and increases at high flow rates. In so far the installation angle of the bend needs to be considered as a parameter for usage.



**Fig. 14: Measuring series at 10D run in distance and different installation angles of the double bend out of plane**

The installation of the double bend out of plane creates even more obvious changes than a simple bend. Because of the swirl the spreading range of the errors raises and the different installation angles lead

to more separated results in the chart. Similar investigation was carried out with a straight input run of 20 D length.



**Fig. 15: Measuring results of 20D straight forward run in distance**

A similarity to the results with 10D run in distance is to observe. Differences are the lower spreading range of the results with 20 D input lengths. Further the results of the perforated plate are more close to the results without straightener.

The following charts show the results of the tests with a bend and a double bend out of plane. Due the longer run in distance the spreading range of the measuring results is smaller in comparison with the 10D tests. The further developed flow profile leads to this smaller spread.

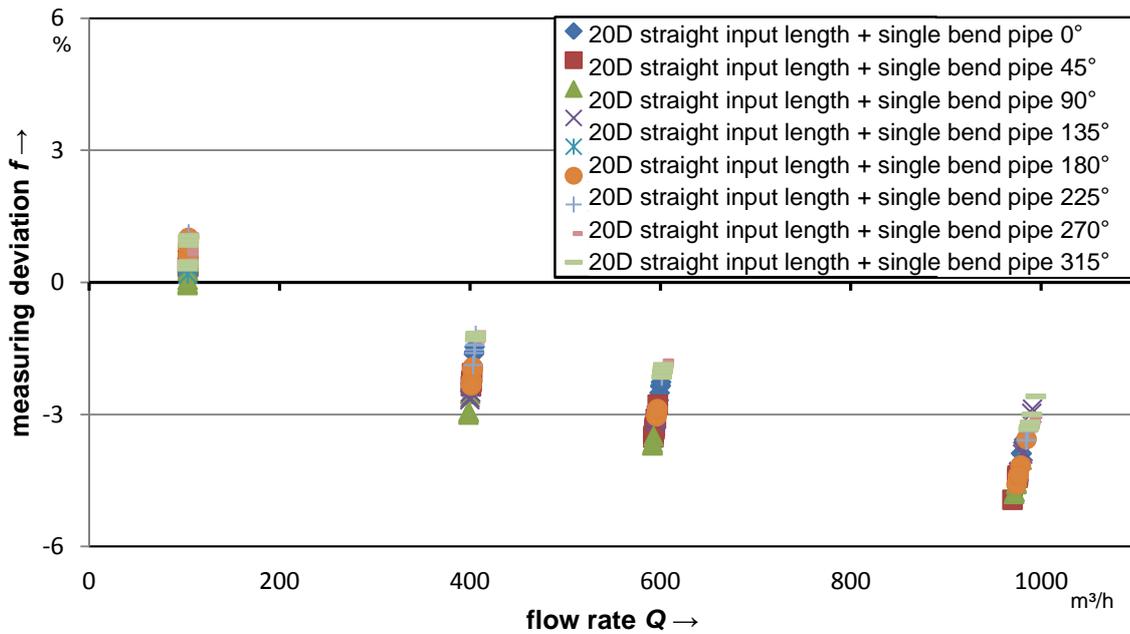


Fig. 16: Measuring results at 10D run in distance and different installation angles of the single bend

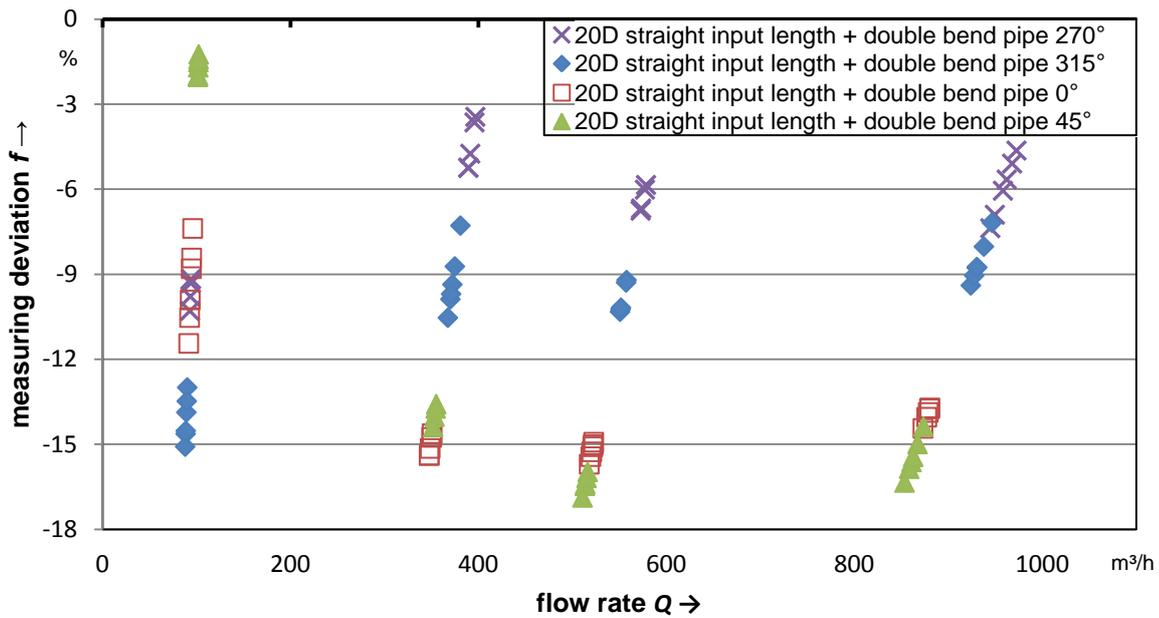


Fig. 17: Measuring results at 20D run in distance and different installation angles of a double bend out of plane

All measuring series demonstrate a dependency on upstream pipe configuration. To reach always a similar developed flow, independently from the

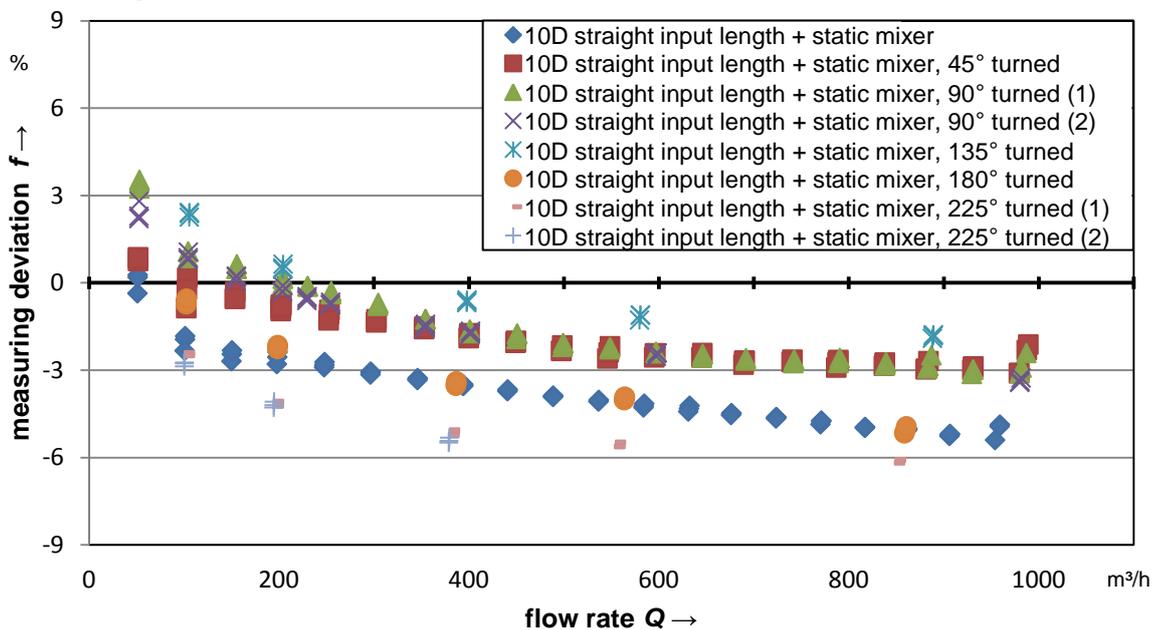
upstream installation, a static mixer located at the inlet of a 10D pipe was investigated.



**Fig. 18: Photograph of the static mixer**

Figure 20 shows the installed mixer. Using the mixer the test procedure described above was repeated. The first test series was focused on the mixer installation angle to check this influence on the result.

This investigation is necessary because of the non axial symmetric design of this kind of flow forming device.



**Fig. 19: Measuring series with static mixer**

For each installation angle a measurement series was carried out. After a test series a turn of 45° clockwise was done.

The results are more repeatable and reproducible and provide good results at high flow rate in comparison to the 0° or 180° installation angle. For the following tests the mixer was installed at the 90°-position.

The best results were observed at the 90° position.



**Fig. 20: Photograph of the installed static mixer at the 45° position and after a 45° turn to the 90° position. The path of the USM is horizontally installed.**

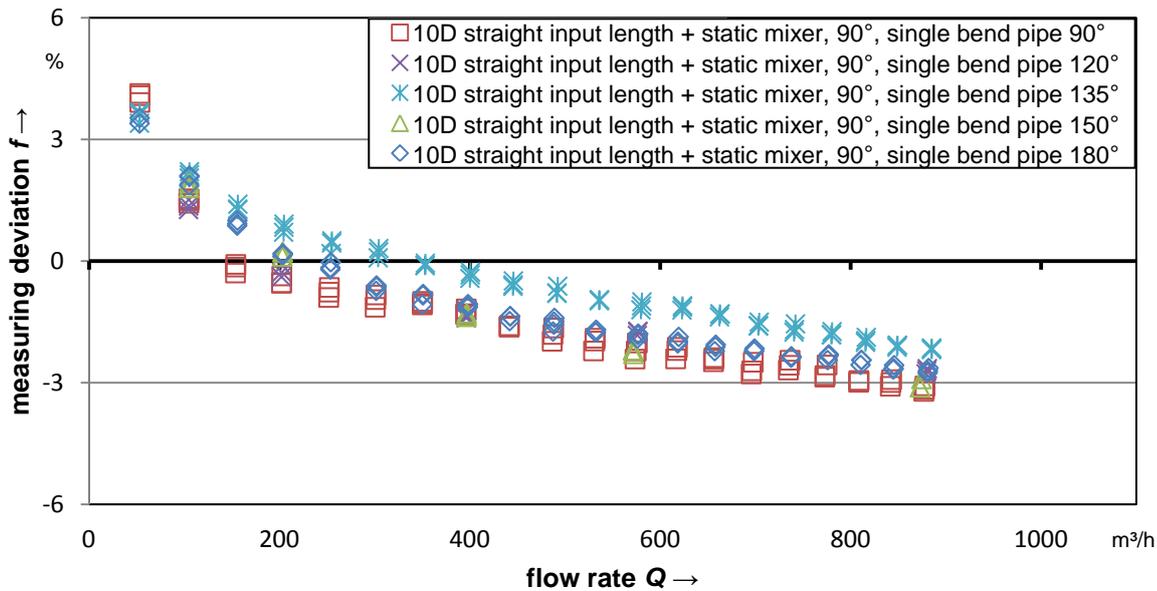


Fig. 21: Measuring results with static mixer at 90° and 10D run in distance and different installation angles of a single bend.

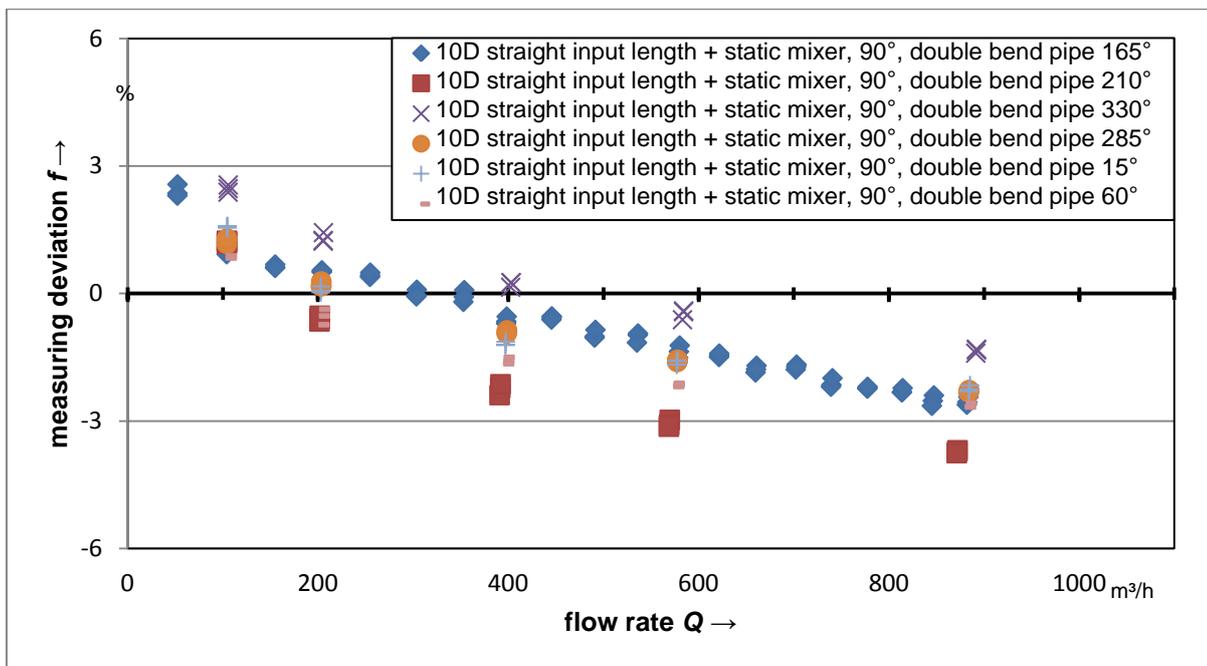


Fig. 22: measuring results with static mixer at 90° and 10D run in distance for different installation angles of the double bend out of plane

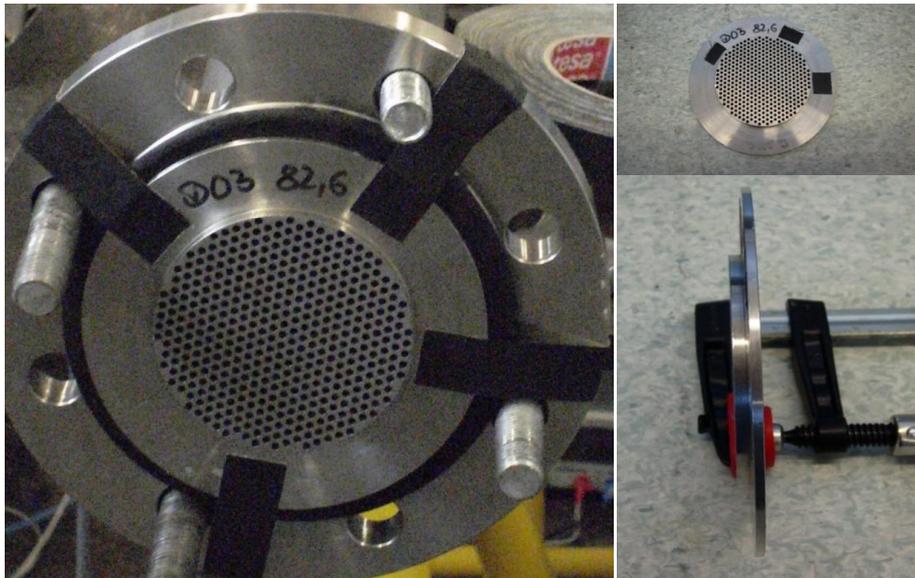
The mixer was installed directly behind the bend so there is no additional run in distance. In comparison to figure 13 the observed error range of the results - especially at the high flow rates - is considerably smaller. Further all results with mixer show a shift to positive errors to observe

The test series carried out with the double bend out of plane show the big advantage of the mixer. The spreading range of the measurement series is relatively small. In addition to the smaller spread the

repeatability is also improved. The results are better than those measured with 20D straight input distance.

It is possible decrease the upstream influences on the results with a static mixer to an extend which allows to reach the 2% uncertainty in the flow determination.

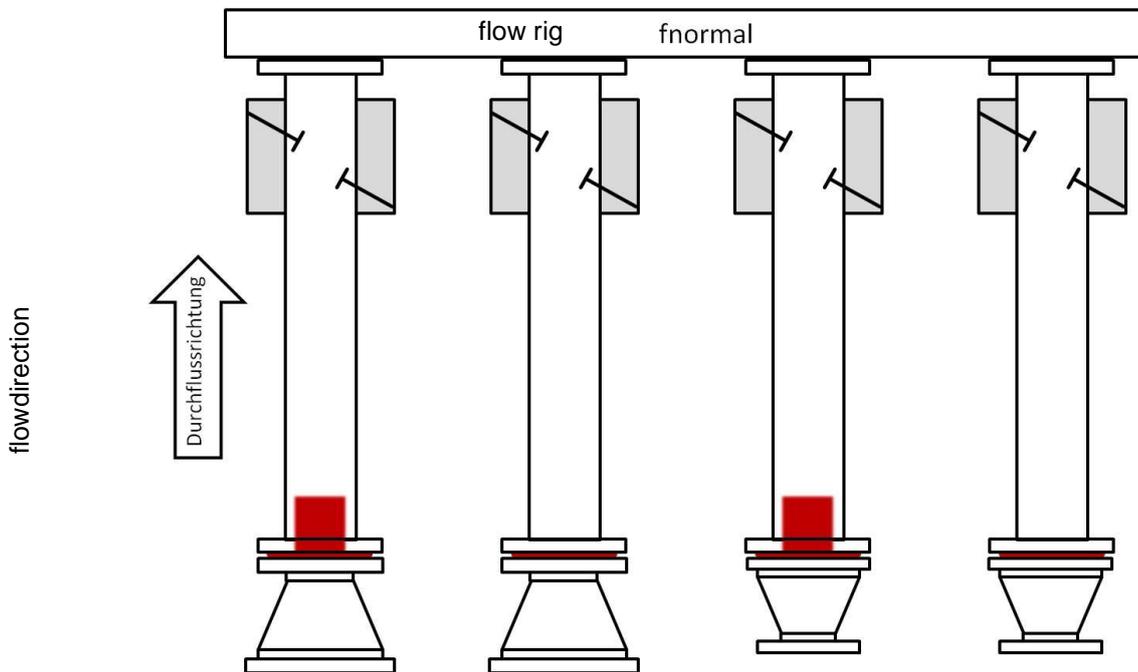
Further a perforated plate was installed at the entrance instead of the mixer.



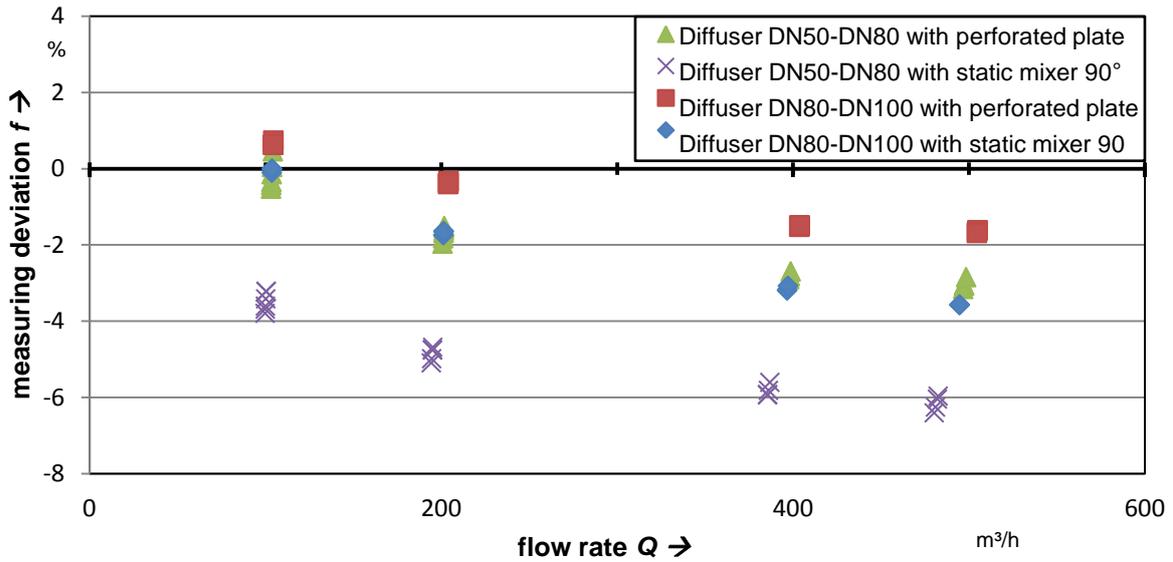
**Fig. 23: Photograph of flow straightener carried out as perforated plate (PTB design).**

Taking into account the application conditions at upgrading facilities to be investigated first in the project the installations described in figure 24 were chosen. Because the sleeve is available for a DN 80 pipe but some of the plants have DN 100 or DN 50 pipes for their exhaust gas these investigations are of interest. A diffuser was placed at the pipe inlet and

outlet as well. Between this diffuser at the inlet and the run in pipe the mixer or the perforated plate was installed and tested respectively. In a second step a DN 100 single bend was installed in front of the bigger diffuser. There were made four measuring series. After each test the bend was turned 45° clockwise.



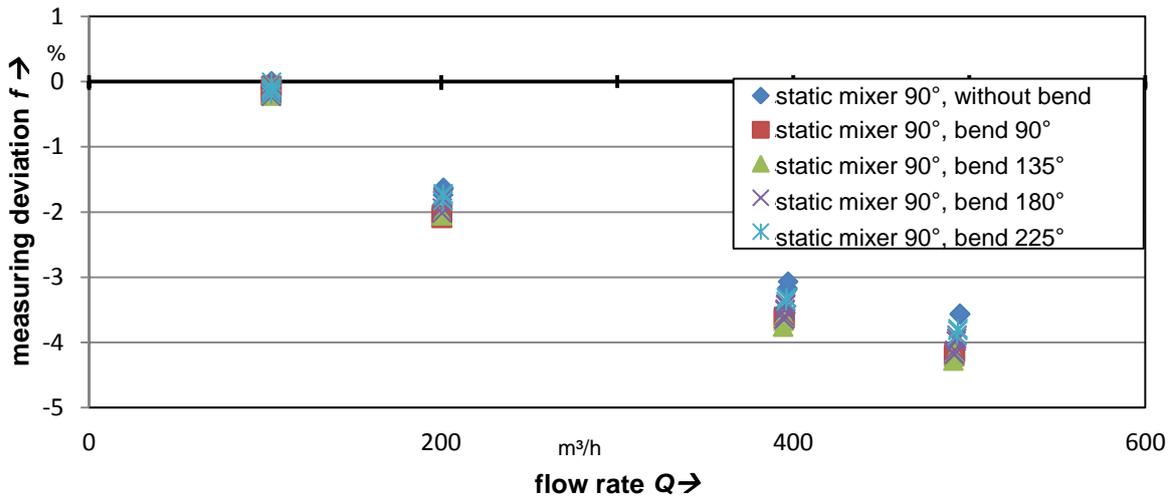
**Fig. 24: Test arrangement for the comparison of static mixer and perforated plate**



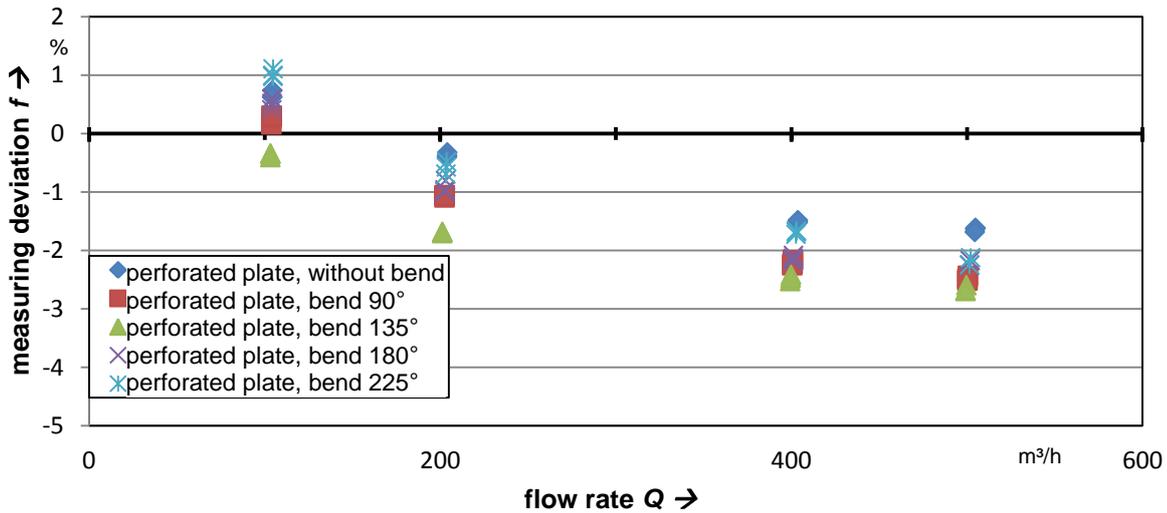
**Fig. 25: Comparison perforated plate and static mixer**

While the results from the test with a DN 80 to DN 100 diffuser have a small spreading range of errors the values from the DN 80 to DN 50 diffuser cover a

large range. It doesn't matter whether the mixer or the perforated plate is used.



**Fig. 26: Static mixer with DN80-DN100 diffuser and single bend at the inlet**



**Fig. 27: Perforated plate with diffuser and single bend**

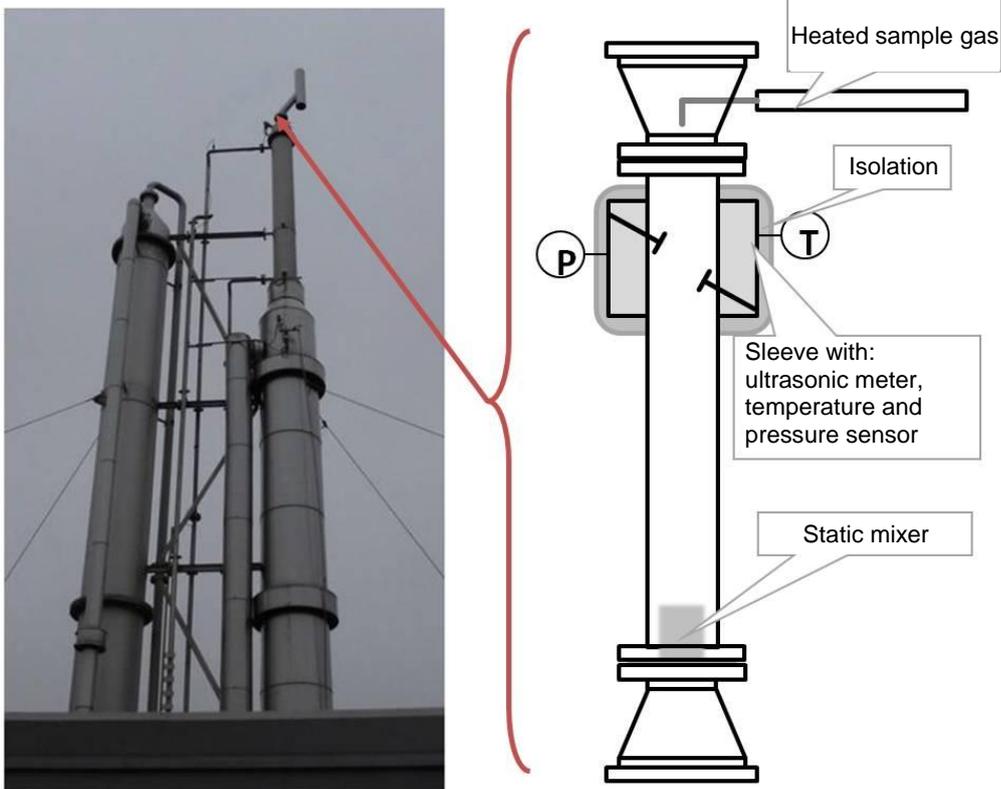
The figures 26 and 27 show, that the results of the measurement series using the mixer are closer together than those with the perforated plate. For applications where the upstream pipe configuration is unknown the mixer should be chosen, to generate a reproducible profile.

## 5 Practical experience

Until now measurement campaigns took place at 2 plants. In the first a scrubbing technique is used.

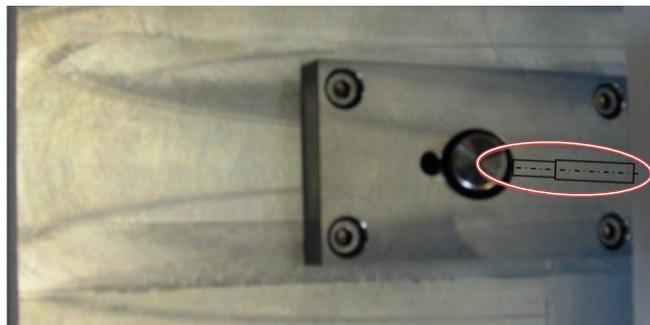
After checking the opportunity to install the flow meter unit it was decided to arrange it very near to head of the exhaust. In this case option 2 shown in figure 4 was applied.

In the exhaust of the facility a flange of DN 100 was available. As shown in figure 28 two diffuser were used to adapt the flow meter module as needed. Figure 28 show a photograph and a schema of the arrangement.



**Fig. 28: Installation of the flow meter module at the upgrading facility**

As mentioned in chapter 2 the exhaust gas of this technique may have very high water vapor content. During the use water condensate flow into the transducer pockets and interrupted the signals. In order to avoid blackouts in flow measurement, a modification of the sleeve was realized to blow out the condensate by pressurized air. Figure 29 show the channel (red marked by an ellipse) where the air was blown in.



**Fig. 29: Arrangement of the blow out channel for the transducer pocket**

It was necessary to blow out the transducer pockets every 3 minutes for up to 5 seconds. After the modification the system worked continuously except during the blow out phase. The carbon dioxide concentration in the exhaust gas was nearly 100 % but this was not critical for the performance values.

The measurement results show concerning the flow rate and in respect to the concentration of methane considerable variations which depend mainly on the load status of the facility. Also relatively large variations with a period of some minutes were observed.

## **6 Conclusion**

During the first two measuring campaigns the exhaust gas measuring system described provided reliable measuring data. As 1 minute average the flow rate measurement data as well as the concentration data are available over a period of 2 weeks. Except during the installation and reinstallation the system worked without personnel on site.

The one path ultrasonic flow metering module could be fitted into the existing piping. As the project is still in progress it is planned to investigate several facilities with other upgrading techniques. In practice there will be many different upstream conditions. Hence further focus has to be on the analysis of the influences of upstream conditions on the meter behavior.

The aim to achieve an uncertainty of  $U = 2\%$  for the volume flow rate determination is reachable if a sufficient input length is available. As far as this is to state now such conditions may be found at the facilities. If a flow conditioner like a gas mixer can be installed upstream to the measurement unit a lower uncertainty is reachable.

## **7 Literature**

- [1] Einspeisung von Biogas in das Erdgasnetz, Fachagentur Nachwachsende Rohstoffe, Leipzig, 2007
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