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Ammonia Emissions: Are Plasma-Physical Techniques Suitable for the Treatment of Exhaust Air from Stalls?

Exhaust air treatment with the aid of non-thermal plasma techniques (NTP) makes it possible to reduce ammonia and odour emissions, as well as the number of germs. In initial tests on a NTP exhaust air cleaning system installed in a pig fattening stall, ammonia concentration was reduced by 18%, however, the energy requirements are significantly higher than for other exhaust air treating systems. Furthermore, the process creates ammonium nitrate, causing further disposal problems.

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Keywords

Non thermal plasma (ntp), ammonia emission, exhaust air treatment

In non-thermal plasma (NTP), the exhaust air is activated by applying a strong electric field so that it becomes a mixture of short-lived radicals and ions which have strongly oxidising properties. Different studies have shown that NTP allows both odours and ammonia to be degraded [1, 2, 3], which also makes its application for emission reduction in animal housing appear possible. The goal of this study was to examine the use of an NTP technique for the treatment of exhaust air from stalls and to develop a pilot system for exhaust air treatment based on the insights gained.

Material and Methods

For the generation of non-thermal plasma (NTP), an NTP reactor (UltraKat company, Gaggenau) with dielectric barrier discharge is used. A strong electric field is applied at two electrodes. A dieletric between the electrodes prevents spark- and arc formation. If high voltage (>1 kV) is applied, a dieletric barrier impedes discharge, and the air which flows through the discharge space changes into the state of plasma (*Fig. 1*).

The plasma-physical exhaust air cleaning system was developed for a fattening pig stall for ten animals. Air from the exhaust air volume flow is blown through the NTP reactor by a centrifugal fan and afterwards fed back into the exhaust air flow (*Fig. 2*). Given high air speed and short residence times of the exhaust air in the NTP reactor, secondary noxious gases, such as laughing gas and nitrous oxides, are avoided. With the aid of this technique, the exhaust air is cleaned either using the flow method, or it is treated with plasma products in the exhaust air shaft using the bypass method.

For gas analysis, an FTIR spectrometer (ThermoNicolet) with a multiple-point sampler was used.

Results and Discussion

The chemical reactions which take place in the discharge space are very complex. The tension activates electrons, which cause the ionisation of the gas mixture due to an avalanche discharge. Direct degradation processes caused by collisions of electrons and ammonia have only little importance in the NTP because these gases only occur in the ppm range and collisions are unlikely [4]. Instead, the energy imparted by collisions with electrons is absorbed by neutral gases, such as nitrogen (70%) and oxygen (21%), of which far larger quantities are present. In the case of oxygen, the imparted energy generates ozone (O_3). Nitrogen contained in the air can be oxidised in the presence of oxygen. As a result, undesirable gases, such as NO,

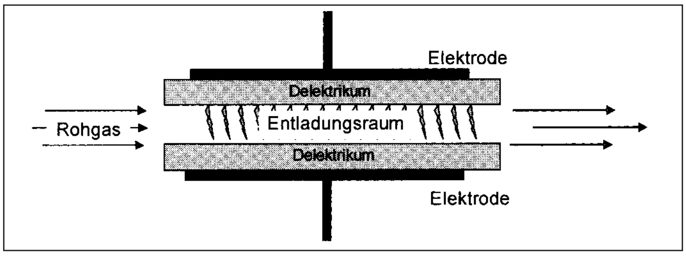

Fig. 1: Unit of a ntp-reactor with dielectric barrier discharge

NO_2, and N_2O, are produced in addition to the activation which generates molecular nitrogen.

With ammonia, several reactions are possible, which can take place simultaneously. For direct ammonia degradation through ionisation, the available energy is too little. For the above-mentioned reasons, the direct reaction caused by collisions with electrons is also likely to have only little importance. Therefore, a large part of the degradation processes generate by-products, such as NO_x and SO_x. In the presence of OH-radicals and NO, these by-products react with ammonia, forming ammonium nitrate (NH_4NO_3) or ammonium sulphate (NH_4SO_4) with nitric acid as an intermediate product.

Direct oxidation of ammonia by ozone to ammonium nitrate is possible as well. The distribution of the mass flows between the products N_2, NH_2, NH_4NO_3, $(NH_4)_2SO_4$, H_2NO_3 is currently still unclear.

The results from the experimental system are shown in *Table 1*. At MP3, the ozone concentration is very high (97 and 238 mg/m^3). For this reason, the exhaust air cleaning system must be run with a residual ozone annihilator. At the current stage of development, ammonia reduction performance must be classified small under the aspect of the energy consumed. Since the measurement series were carried out on different days, the efficiency of NH_3 reduction is based on the calculated NH_3 load. Irrespective of the power applied at the NTP reactor, ammonia concentration in the stall air (6.6 and 7.1 ppm) at MP3 was reduced by 18%. If the reaction path was shorter (MP2), efficiency was considerably lower.

As compared with MP1 and MP3, nitrous oxide concentration at MP2 is higher at both power stages. This confirms the hypothesis that nitrous oxide is a product of plasmagenesis and a reaction partner for ammonia after reaction with water vapour to HNO_3.

Based on the data material, no directed effect can be discerned in laughing gas concentrations. An increase in N_2O concentration due to NTP treatment cannot be observed.

Based on a laboratory trial carried out beforehand [5], considerably higher efficiency was expected. This may be caused by the ventilation cross section, which was significantly larger as compared with the laboratory trial. As a result, it is less likely that an NTP radical and an ammonia molecule meet. Therefore, it seems appropriate to combine NTP exhaust air cleaning with other methods in order to annihilate the large contents of residual ozone and to increase the conversion rates by enlarging the reaction surface.

Currently, great insecurity still lies in the detection of nitrogenous by-products, such as ammonium nitrate. Like saltpetre, the latter cannot be detected with the aid of FTIR spectroscopy. Therefore, additional analytical methods must be developed. Due to the short trial interval and the large surface, no deposits of ammonium nitrate were able to be found in the exhaust air pipes.

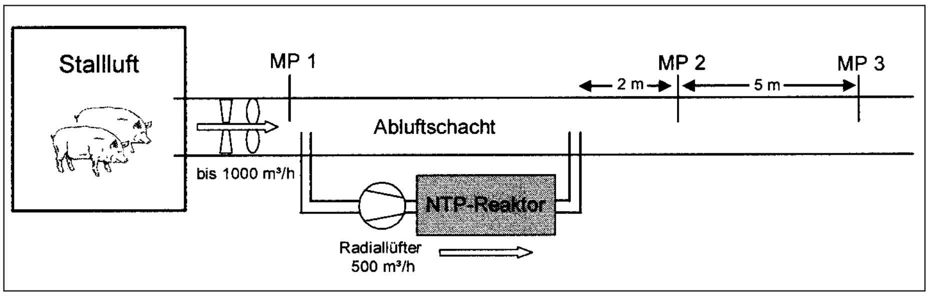

Fig 2: Schematic diagram of a waste air treatment facility with a ntp-reactor and the measuring points (MP) for gas analysis in an experimental stable for ten fattening pigs

Conclusions

With regard to the reduction of ammonia emissions from animal housing, exhaust air treatment based on a plasma-physical technique can currently only be applied under certain conditions. As compared with other methods of exhaust air treatment, efficiency is low, and the occurrence of new degradation products, such as ammonium nitrate, raises new questions as to their disposal and utilisation. Nevertheless, combinations with other techniques of exhaust air treatment should be examined which include odour emissions and germ reduction in addition to the problems caused by ammonia.

NTP [W]	MP	Air vol. flow [m^3/h]	O_3 [mg/m^3]	N_2O [mg/m^3]	NO_X [mg/m^3]	NH_3 [mg/m^3]	NH_3 WG*
1250	1	818	0	1,41	0,98	6,60	
	2	818	99	0,95	3,82	6,17	6%
	3	818	97	1,65	1,88	5,39	18%
2500	1	645	8	2,12	0,35	7,11	
	2	651	238	0,12	1,79	6,54	7%
	3	645	238	0	0,47	5,84	18%

* Efficiency is based on the calculated NH_3 loads.

Table 1: Effect of ntp-waste air treatment in pig fattening stable (10 fattening places) on nitrogenous gas compounds and the efficiency of NH3 reduction (WG), dependent on ntp-capacity (1250 W; 2500 W) and the reaction path (MP1 = raw gas; MP2 , MP3= purified gas

Literature

[1] *Zhang, R., T. Yamamoto and D. S. Bundy*: Control of Ammonia and Odors in Animal Houses by a Ferroelectric Plasma Reactor. In: IEEE Transactions on Industry Applications 32 (1996), no. 1, pp. 113-117

[2] *Wang, Y., and R. Goodrich*: Destruction of Swine Manure Gases by a Non-Thermal Plasma Reactor. In: International Symposium on Gaseous and Odour Emissions from Animal Production Facilities. EurAgEng, Horsens, 1.-4. Juni 2003, pp. 444-451

[3] *Rice, R.G.*: Ozone for Air Treatment - Basic Principles. In: International Ozone Association, 16th World Congress, 31. 8-5. 9. 2003, Las Vegas, pp. 1-23

[4] *Hongbin, M., P. Chen and R. Ruan*: H_2S and NH_3 Removal by silent discharge Plasma and ozone Combo system. Plasma Chemistry and Plasma processing, 21 (2001), no. 4

[5] *Kosch, R., A. Decker and H. Van den Weghe*: Application of non-thermal plasma techniques (NTP) to reduce emissions from animal husbandry. International Congress of Animal production in Europe: The way forward in a changing world, ISAH, Saint Malo, 11.-13. Oct. 2004, pp.67-68